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Self-optimization of distillation sequences using DSE parameter.

Auto-optimització de trens de columnes de destil·lació mitjançant la eficiència de la seqüència de destil·lació (DSE)

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Hi ha una força motriu més poderosa que el vapor,
l'electricitat i l'energia atòmica: la voluntat.

Albert Einstein

REPORT

CONTENTS

1. SUMMARY

To perform an azeotropic ternary mixture separation, several distillation sequence are feasible to separate the three compounds of the mixture. Due to the great complexity of the process the mathematical model is simplified according to the ∞/∞ analysis assumptions, i.e. columns of infinite length operated at the infinite reflux.

The objective function is the overall process energy requirements that in previous studies was approximated to the overall process distillate flow rate. The problem of this optimization function is that it does not take into account the facility or difficulty to perform this separation and the efficiency of the columns.

The originality of the present study is that the distillation sequence efficiency (DSE) of the overall process is used as the optimization function. The energy requirement depends on the crude feed composition. The novel method is applied to a case study for the separation of a crude feed formed by methanol, 2-propanol and water.

The aim of this project is to compare the results previously obtained by Ulrich and Morari (2003) minimizing the overall distillate flow rate with the results maximizing the DSE, i.e. the efficiency of the columns is taken into account.

Keywords: ∞/∞ analysis, column sequencing, heuristics, boundary line, azeotrope

2. RESUM

Per dur a terme una separació d'una mescla ternària azeotròpica, varies seqüències de destil·lació són factibles per separar els tres compostos de la mescla. A causa de la gran complexitat del procés, el model matemàtic es simplificat mitjançant les suposicions de l'anàlisi ∞/∞, és a dir columnes d'infinita altura i operades a infinit reflux.

La funció objectiu és els requeriments d'energia de tot el procés que en previs estudis va ser aproximada als cabals totals de destil·lat. El problema d'aquesta funció d'optimització és que no té en compte la facilitat o dificultat per dur a terme aquesta separació i la eficiència de les columnes.

La originalitat del present estudi és que la eficiència de la seqüència de destil·lació (DSE) del procés global sí que és utilitzada com a funció d'optimització. El requeriment d'energia depèn de la composició de l'aliment cru. El nou mètode es aplicat a un cas d'estudi per la separació d'un aliment cru format per metanol, 2-propanol i aigua.

L'objectiu d'aquest projecte és el de comparar els resultats anterior obtinguts per en Ulrich i en Morari (2003) minimitzant el cabal global de destil·lat amb els resultats maximitzant el DSE, es a dir la eficiència de les columnes es té en compte.

Paraules clau: Anàlisi ∞/∞, seqüenciació de columnes, heurístics, línia de frontera, azeòtrop

3. INTRODUCTION

Distillation is the most used unitary operation to separate liquid mixtures. When a mixture is zeotropic, its separation is performed with a simple distillation sequence but, if the mixture is azeotropic, the separation is not so easy and enhanced distillations such as extractive or heterogeneous azeotropic distillation is required.

To achieve a multicomponent separation, a feasibility analysis is required to choose and design feasible sequences able to perform the desired separation.

3.1. ANALYZED PROBLEM

Ulrich and Morari (2003) analyzed a feasible sequence to separate a ternary azeotropic mixture of methanol, 2-propanol and water. The distillation sequence proposed was formed by three distillation columns with two recycles of the distillates of columns 2 and 3.

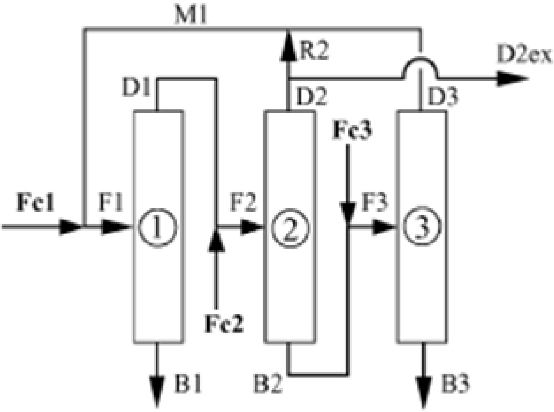

Figure 1: Distillation sequence proposed by Jan Ulrich and Manfred Morari (2003)

As shown in Figure 1, the ternary mixture is fed to the first column mixed previously with the recycling stream of columns 2 and 3 (M_1). In the column 1, part of the water is separated from the mixture in the bottom of the column (B_1) and at the top of the column (D_1) the ternary mixture is collected. This stream is fed to the second column, where the light compound (methanol) is collected at the top of the column (D_2).

Besides distillate flow rate, (R_2) is recirculated. From the bottom of the column 2, the binary mixture water/2-propanol stream is fed to the third column where 2-propanol (B_3) is collected at the bottoms and the azeotropic mixture water/2-propanol (D_3) is collected at the distillate and mixed with R_2 to produce the stream M1.

To simplify the complex resolution of this distillation column train, Ulrich and Morari (2003) used the ∞/∞ analysis in which it's supposed that the height of the column and the reflux rate are infinite, which makes that the column profile includes a singular point (pure compounds or azeotropes).

Ulrich and Morari (2003) proposed a self-optimization procedure providing the possibility that the crude feed, depending on its composition, can entry in one column or another. The procedure has a great novelty because it is able to provide the suitable distillation sequence depending on the crude feed composition instead of fixing the crude feed composition as done in most of the studies nowadays available in open literature.

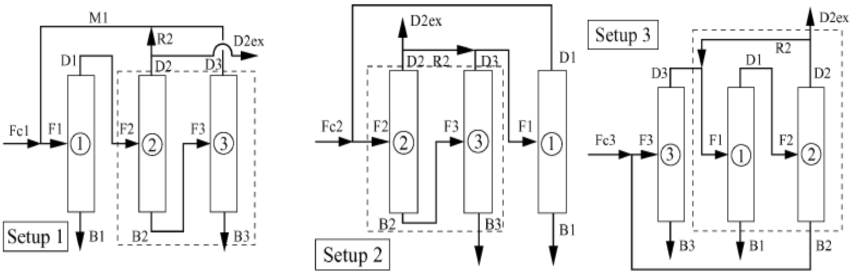

Figure 2: Three setups proposed by Jan Ulrich and Manfred Morari (2003)

3.2. OPTIMISATION PROBLEM

To optimise de distillation sequence, a cost function must be used. Ulrich and Morari (2003) proposed the sum of the distillate flow rates of the three columns as the cost function.

To optimize the distillation sequence, the variables used to fulfil the available degrees of freedom are chosen. Distillation is very energy intensive and the overall operation costs are proportional to the overall reboiler duty requirements, in this case of the three columns (Equation 1).

$$J^D = Q_1 + Q_2 + Q_3 \qquad (1)$$

Disregarding the energy related to the reflux, the reboiler duty is proportional to the distillate flow rate; being the proportionality constant, its heat of vaporization (Equation 2).

$$Q_i^R = p_i(r_i + 1)D_i = k_i D_i \qquad (2)$$

The authors developed a cost function made of the three distillate flow rates. When the distillate flows rates are higher, the reboilers duties are higher the same than the operation. Under these assumptions, a simple cost function is proposed (Equation 3).

$$J^D = k_1 D_1 + k_2 D_2 + k_3 D_3 \qquad (3)$$

The k_i values of finite columns depend on the mixture and on the column design. For a sequence of ∞/∞ columns, which will be investigated here using the residue curve boundary, a further approximation is made: $k_1=k_2=k_3=k_i$. This assumption is derived from considering a constant heat of vaporization, which is performed in many simplified methods for distillation such as the McCabe-Thiele. In this case, J is minimum if the sum of the distillate flow rates is minimum (Ulrich and Morari, 2003) (Equation 4).

$$J^D = D_1 + D_2 + D_3 \qquad (4)$$

To optimize this cost function, it is expressed in function of more suitable variables as is discussed in the following paragraph using the mass balance in column 1 and $D_2 = R_2 + D_2^{ex}$, (Equation 5).

$$J^D = 2(R_2 + D_3) + F_c - B_1 + D_2^{ex} \qquad (5)$$

The system has two degrees of freedom (see point 5.3. for a brief discussion or Ulrich and Morari (2003) for a detailed demonstration). F_c is the feed flow rate and B_1 and D_2^{ex} are fixed by the feed and the overall process mass balance. Ulrich and Morari (2003) chose R_2 and D_3 to optimise this function. Minimizing these two flow rates, the minimum operation costs are attained according to the optimization function.

In Figure 3, D_3 is minimum when x^{D3} is in the I/H azeotrop, for this reason, D_3 is fixed and we can get the optimal R_2 to give the minimum J^D.

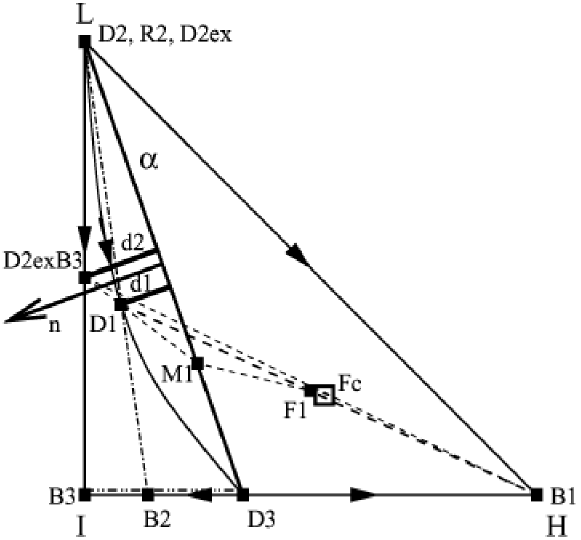

Figure 3: Residue curve diagram

For the lever rule (Ulrich and Morari 2003):

$$\frac{R_2 + D_3}{D_2^{ex} + B_3} = \frac{d_2 - d_1}{d_1} \qquad (6)$$

As shown in Figure 3, d_1 and d_2 represent the distances between the two composition x^{D1} and $x^{xD2exB3}$ and the line R that connects x^{D2} with x^{D3}. Eq. 6 indicates that $R_2 + D_3$ is minimized when d_1 is maximized (Ulrich and Morari, 2003). For this reason there is a $x^{D1,\ opt.}$ optimal in the boundary line that maximize d_1.

The optimal J^D is proportional to $(1 - x_H^{Fc} – x_L^{Fc})$ (Ulrich and Morari, 2003). In particular, for a $x^{D1,opt}$ [molar fraction: 0.25 of H, 0.375 of L and 0,375 of I] and for a $x^{D3,opt}$ [molar fraction: 0.5 of H and 0.5 of I], J^D for Setup 1is presented in equation 7 and for Setup 2 in equation 8:

$$J^D = 15(1 - x_H^{Fc} - x_L^{Fc})F_c \qquad (7)$$

For Setup 2:

$$J^D = (17x_H^{Fc} + x_L^{Fc})F_c \qquad (8)$$

In Setup 3 J^D is not a linear function of x^{Fc}. In this sequence the optimal values of x^{D1} and x^{D3} depend on the feed composition.

Figure 4 shows the cost function results for the three Setups versus the crude feed composition.

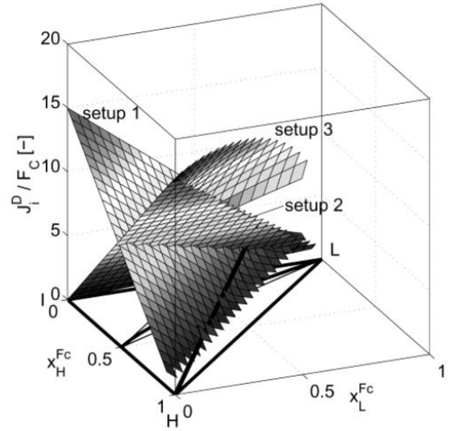

Figure 4: Optimisation graphics of (Ulrich and Morari, 2003)

Heuristics are based on experience and are commonly used to choose the best setup of a distillation sequence. However they have the limitations derived from the fact that they are applied sequentially to each separation and not to the overall distillation system. Therefore, it is not easy to apply heuristics to a distillation sequence with recycle streams and furthermore, sometimes they contradict each other. CDS and CES are introduced to help when choosing the best setup is not clear from the heuristics.

Table 1 lists the main heuristics rules and evaluation functions that have been proposed to date for the design of distillation sequences. In the absence of recycling streams, the method proposed by Ulrich and Morari (2003) minimizing the distillate flow rate is equivalent to the heuristic favouring the direct sequence.

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Table 1: List of the main heuristics

Heuristic	Reference
Perform the easiest separation first	[Harbert 57] [Rudder al 73]
Remove the most plentiful component first	[Nishimura & Hiraizumi 71] [King 71] [Rudder al 73]
Favour the 50/50 split	[Harbert 57] [Heaven 69] [King 71]
Perform the cheapest separation first	[Harbert 57] [Rudder al 73]
Perform the separation with the smallest CDS first	[Nath & Motard 81]
Perform the separation with smallest CES	[Liu 87]
Perform the separation with smallest energy-index first	[Lien 88]
Favour the direct sequence	[King 71]
Favour a separation in the absence of non-key component	[Heaven 69] [King 71] [Gomez & Seader 76]

Although the use of heuristic rules is computationally cheaper than conducting a search, the quality of the decision taken and that of the final solution obtained, may not be the best choice.

4. OBJECTIVES

The aim of this project are:

- Check the suitability of DSE as optimisation function to optimise a ternary mixture distillation sequence.

- Develop a calculation program in Matlab® that provides the most efficient Setup for any crude feed composition.

- Develop a post processing program in Matlab® to graphically represent the results to identify the ranges of composition in which the Setups are most efficient and make a comparison between the three Setups.

- Discuss critically the results using the Ulrich and Morari (2003) optimisation function or the DSE function.

5. METHODOLOGY

This section describes the software tools used in this study for simulation and calculations, as well as the assumptions and the methodology used.

5.1. SOFTWARE TOOLS

The thermophysical properties of the three separation systems are simulated using Simulis Thermodynamics®. It is a calculation server for thermophysical properties and phase equilibria calculations on pure components and mixtures. It is available as a Microsoft Excel® add-in, a toolbox in MATLAB® or as a software component which can be easily plugged in any other CAPE open application requiring reliable and accurate thermophysical properties.

Material balances, graphics, DSE yields and optimal values are calculated using MATLAB® V7.7.0.471 (R2008b) (The MathWorks, Inc.). It is a multi-paradigm numerical computing environment and fourth-generation programming language.

5.2. GENERAL SYSTEM CHARACTERISTICS

The compounds considered in this system are:

- Methanol: Light compound (L)
- 2-Propanol: Intermediate compound (I)
- Water: Heavy compound (H)

A calculation basis of 1,000 kmol/h is used in the mass balances of all the Setups. Also is used the ∞/∞ analysis to simplify the calculations, for this reason all column profiles contain at least one singular point. The feed is separated into pure compounds which are collected at the system output streams and multicomponent mixtures are feed to the columns. The presence of azeotropes leads to the need to use recycle streams.

The separation is performed by different distillation train configurations. Figure 1 shows one feasible Setup called in this study Setup 1. Column 1 separates the heavy compound (H) at the bottom of the column and ternary mixture is extracted at the top of the column. Column 2 extracts at the top all the light compound (L) and at the bottom a binary mixture of the heavy (H) and the intermediate (I) compounds. Finally, column 3 collects at the bottom the intermediate compound and at the top of the column the binary mixture of H/I compound. Changing the crude feed column, all the feasible separation schemes are generated.

5.3. DEGREES OF FREEDOM OF THE ∞/∞ SEQUENCE

Because of the complexity of the process with two recycles, the analysis starts with three columns in a row.

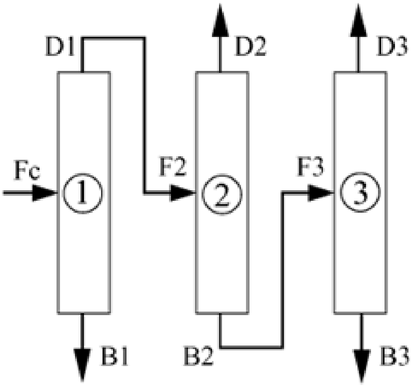


Figure 5: Three columns in a row with any recycle

A distillation column separating a *c*-component mixture has *c + 2* degrees of freedom according to the ∞/∞ analysis assumptions. Hence, a three-column sequence has 3c + 6 degrees of freedom. For a given external crude feed Fc, *c* degrees of freedom are fixed: *c – 1* for the feed composition and one for the feed flow rate. In addition, *2c* degrees of freedom are fixed by the selected connections: $D_1 = F_2$ and $B_2 = F_3$. Assuming that the columns are operated at atmospheric pressure, three more degrees of freedom are fixed. Hence, *3c + 3* degrees of freedom are fixed, leaving three degrees of freedom for the sequence. By specifying B_1, D_2 and

D_3, all flow rates and product compositions are determinate. In similar way, when the B_1, D_2 and D_3 output streams compositions are fixed to pure products then all the system becomes defined without remaining any degree of freedom.

Figure 6 shows the three-column sequence with D_3 recycled to the feed of column 1. Each compound trapped in the system add one degree of freedom, therefore due to the recycle present has one degree of freedom.

In similar way, with two recycled streams (Figure 2) then two degrees of freedom are present. Therefore assuming that the crude feed stream variables are fixed and that pure compounds are collected by B_1, D_2 and D_3, then the overall process has two degrees of freedom according to the ∞/∞ analysis.

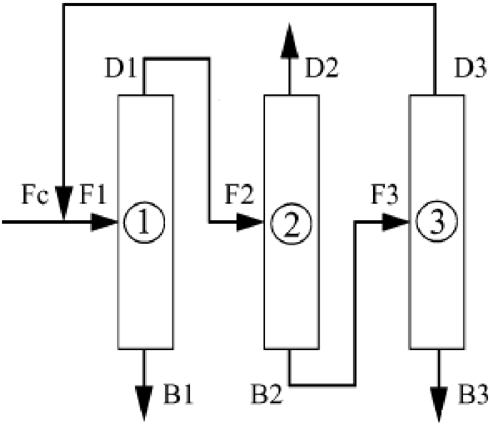


Figure 6: Three columns with one recycle (D_3)

5.4. METHODOLOGY

5.4.1. Setup 1

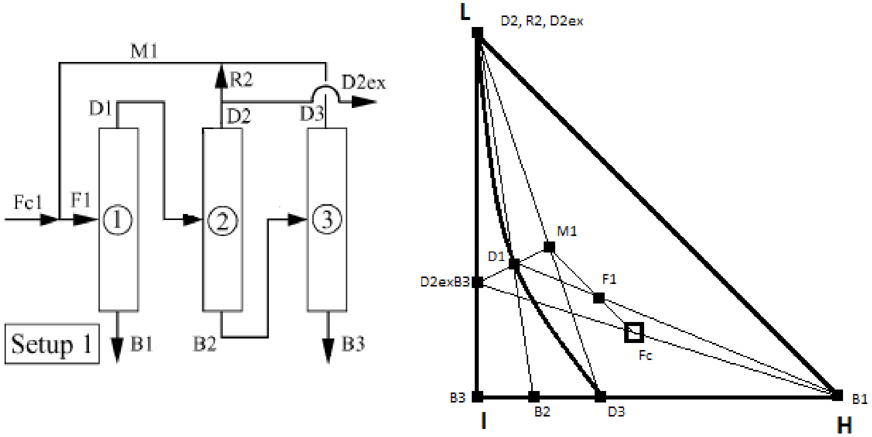


Figure 7: Setup 1 sequence and residue curve diagram

5.4.1.1. Firsts Assumptions

First of all, all the streams, flow rates and the compositions of the system are calculated. Once the crude feed composition is defined:

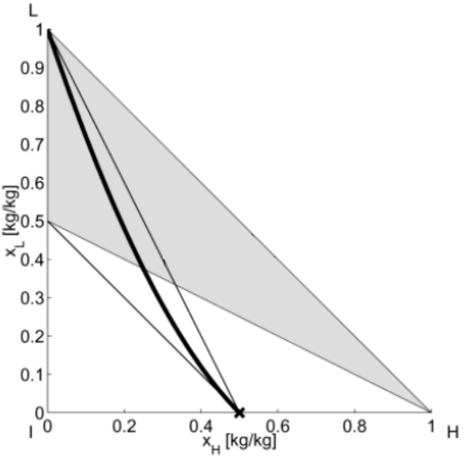
$$x_H^{Fc} = 0{,}3 \;\; ; \;\; x_L^{Fc} = 0{,}3 \;\; ; \;\; x_I^{Fc} = 0{,}4 \;\; ; \quad Fc = 1{,}000\, {}^{kmol}/_{h} \qquad (9)$$

According to the ∞/∞ analysis, pure compounds are collected at output streams so, $x_H^{B1} = 1$, $x_L^{D2ex} = 1$ and $x_I^{B3} = 1$.

Hence, streams B_1, D_2^{ex} and B_3 are calculated by the overall mass balance once fixed the feed stream. These streams are not dependant on any other variables.

At the top of column 2, the light compound is collected therefore, $x_L^{D2ex} = x_L^{R2} = 1$.

As can be seen in the residue curve diagram, the composition of D_1 is located on the boundary. The boundary line is fit to a quadratic function (Equation 10), to the model implemented in the program. The boundary line is between the azeotrope $x_H^{D3} = x_I^{D3} = 0{,}5$ and pure light compound (Figure 8).



$$x_L^{Wi} = 2(x_H^{Wi})^2 - 3x_H^{Wi} + 1 \quad (10)$$

Figure 8: Residue curve diagram approximated to a quadratic function

5.4.1.2. Optimisation function

A good optimization function is the reboiler duty required by the three distillation columns simulated using a rigorous model. A rigorous model simulation to evaluate a large number of alternatives is impractical from the point of view of calculation power and convergence difficulties. For this reason, Ulrich and Morari (2003) use as optimization objective function the overall distillate flow rate, assuming that the energy is proportional to the distillate streams. Unfortunately, the difficulty to perform each separation is not taken into account under this assumption. The goal of the present study is to perform the optimization, but taking into account the efficiency of the columns. Higher distillation sequence efficiency is obtained for high DSE parameter values close to one.

DSE is a parameter useful to determine how much efficient is a distillation sequence. It takes into account the molar fraction of the compounds in the feed stream and the Carnot efficiency of the column. As many streams are closed in the system, the DSE is calculated from the sum of efficiencies of output and recycling streams flow rate divided by the crude feed flow rate.

The efficiency depends on the path followed by the stream. An output stream efficiency depends on the columns efficiency from its path that is form feed and for the recycles the path forward is from the point where the recycled is mixed to where is recycled back and the path backwards is the recycle itself. The recycle flow rate has a negative efficiency of one for the path backwards.

The DSE definition for a sharp split sequences is presented in equation 11.

$$DSE = \sum_i x_i \cdot \prod_C \eta_{iC} \qquad (11)$$

where, X_i = molar fraction of the compound in the feed of the sequence.

η_{iC} = Carnot efficiency of the column C.

In this work, general DSE presented in equation 12 is used.

$$DSE = \sum_i W_i \cdot \prod_C \eta_{iC} \qquad (12)$$

where, W_i = stream depending on the feed.

η_{iC} = Carnot efficiency of the column C

The Carnot efficiency of the three columns are presented in equation 13 to 15.

$$\eta_1 = \frac{(T_{B1} - T_{D1})}{T_{B1}} \quad (13)\,; \quad \eta_2 = \frac{(T_{B2} - T_{D2})}{T_{B2}} \quad (14)\,; \quad \eta_3 = \frac{(T_{B3} - T_{D3})}{T_{B3}} \quad (15)$$

The Carnot efficiency is applied to the distillates and at the bottom, the efficiency is 1.

For Setup 1, accordingly to the last considerations, the optimisation function is presented in equation 16.

$$DSE_1 = \frac{B_1}{Fc} + \frac{B_3}{Fc} \cdot \eta_1 + \frac{D_2^{ex}}{Fc} \cdot \eta_1 \cdot \eta_2 + \frac{D_3}{Fc} \cdot (\eta_1 \cdot \eta_3 - 1) + \frac{R_2}{Fc} \cdot (\eta_1 \cdot \eta_2 - 1) \qquad (16)$$

The efficiency of the sequence is maximum when the DSE parameter is maximized. As B_1, B_3 and D_2^{ex} become fixed by the overall mass balance, the variables used as to fulfil the two degrees of freedom available are D_3 and R_2.

5.4.1.3. Methodology for Setup 1

As the sequence has two degrees of freedom (see section 5.3), D_3 and R_2 are chosen as degrees of freedom of the system. The objective is to develop a program code to find the values of D_3 and R_2 that maximize the DSE.

First, the rest of mass balances are implemented using as input data the calculation crude feed stream and D_3 and R_2 flow rates (process manipulated variables) (see Appendix 1).

A bisection and Bolzano theorems are implemented in MATLAB® to obtain the manipulated variable values that maximizes the objective function.

To speed up the optimization the composition of D_1 is located on the boundary line (Figure 7).

The boundary line is implemented in the program as a quadratic function (equation 17). x_L^{D1} must satisfy the condition that the light compound composition in column 1 calculated from the mass balances must be the same than calculated with the boundary line quadratic function (eq. 18). This condition places column 1 distillate composition on the boundary line.

$$x_L^{D1} = 2(x_H^{D1})^2 - 3x_H^{D1} + 1 \qquad (17)$$

Therefore,

$$x_L^{D1} = x_{Lbl}^{D1} \qquad (18)$$

As a summary, once implemented the mass balances then Bolzano theorem is used to optimize D_3 together with bisection theorem to optimize R_2 fulfilling that x_L^{D1} is located on the boundary line. The procedure is repeated thousands of times to determinate the DSE for all feed compositions regions.

5.4.2. Setup 2

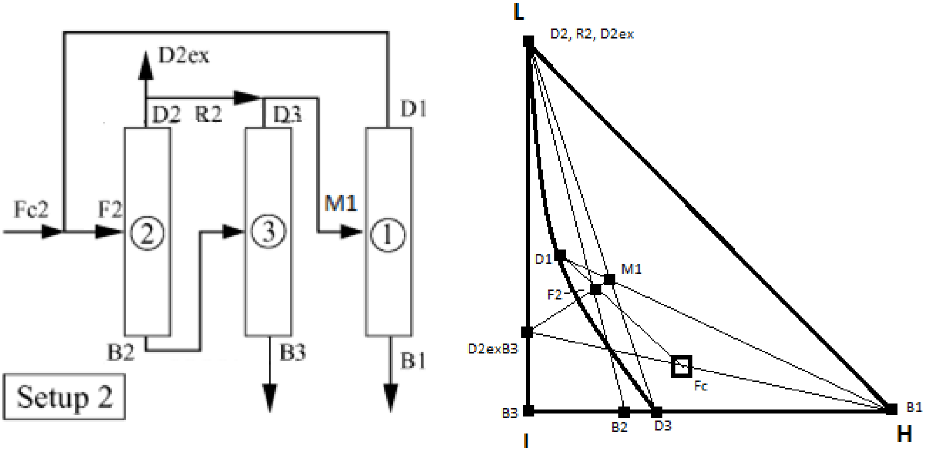


Figure 9: Setup 2 sequence and residue curve diagram

5.4.2.1. Firsts assumptions

Setup 2 assumptions are the same that in Setup 1. All the output streams are pure components and degrees of freedom are 2. As Setup 1, the variables used as to fulfil the two degrees of freedom available are D_3 and R_2. In this sequence there is only one recirculation back (D_1) and a part of D_2 is not collected but fed to the third column, hence the optimization function is different than in Setup 1.

5.4.2.2. Optimization function

The DSE function will depend on output streams and recirculation streams. D_1 is the only recirculation stream in this sequence as Figure 9, but a part of D_2 called R_2 is reintroduced in the system.

R_2 is mixed with D_3 to form M_1 and then it pass to form part of D_1 because column 1 only separates de heavy compound of the others hence it forms part of D_1 and is recycled.

$$DSE_2 = \frac{D_2^{ex}}{Fc} \cdot \eta_2 + \frac{B_3}{Fc} + \frac{B_1}{Fc} \cdot \eta_3 + \frac{D_1}{Fc} \cdot (\eta_3 \cdot \eta_1 - 1) + \frac{R_2}{Fc} \cdot (\eta_2 \cdot \eta_1 - \eta_3 \cdot \eta_1) \qquad (19)$$

As it can be seen in equation 19, in contradiction to Setup 1 (where parameters that are used to optimise DSE form part of the DSE function), D_3 does not form part of the optimisation function but D_1 depends on D_3 hence it is preferred to perform the optimisation with the same manipulated variables as of Setup 1.

5.4.2.3. Methodology for Setup 2

As in Setup 1, the same parameters are maximized so the same methodology is used. The intermediate condition is the same (Figure 9). D_1 is in the boundary line hence x_L^{D1} must satisfy the quadratic approximation of the boundary line.

$$x_L^{D1} = x_{Lbl}^{D1} \qquad (20)$$

The optimisation process is applied to all the range of crude feed compositions.

5.4.3. Setup 3

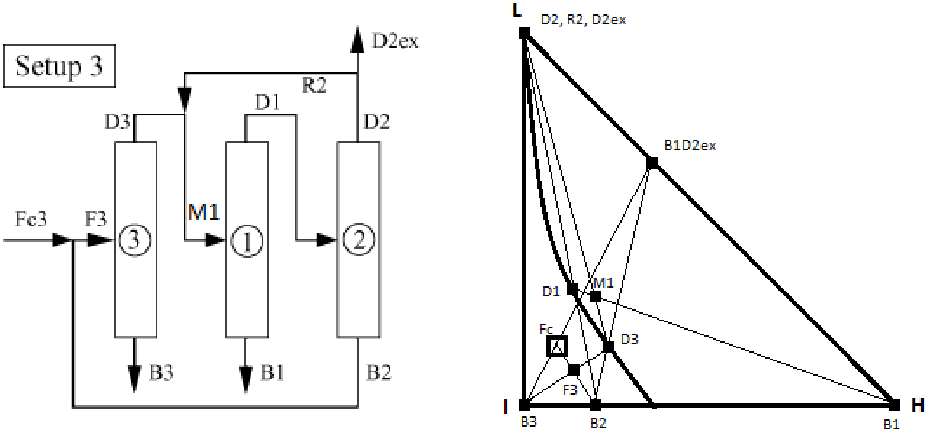


Figure 10: Setup 3 and residue curve diagram

5.4.3.1. First assumptions

All the composition and streams are determinated as for Setup 1 and Setup 2. Initial assumptions are the same than for previous Setups but, D_3 is not located in the boundary hence its composition is not fixed.

According to the ∞/∞ analysis, pure compounds can be achieved in the output streams hence, $x_L^{D2ex} = x_H^{B1} = x_I^{B3} = 1$.

5.4.3.2. Optimisation function

The optimisation function of the DSE (with the same Carnot efficiency for the three columns as in Setup 1 and 2) is different because in this Setup, D_3 is not a recirculation stream and does not appear in the optimisation function. In this Setup, B_2 is the recirculation stream that is taken into account in the DSE function (equation 21).

$$DSE_3 = \frac{D_2^{ex}}{Fc} \cdot \eta_3 \cdot \eta_1 \cdot \eta_2 + \frac{B_3}{Fc} + \frac{B_1}{Fc} \cdot \eta_3 + \frac{B_2}{Fc} \cdot (\eta_3 \cdot \eta_1 - 1) + \frac{R_2}{Fc} \cdot (\eta_1 \cdot \eta_2 - 1) \quad (21)$$

5.4.3.3. Methodology for Setup 3

The sequence has two degrees of freedom (see section 5.3), the sequence is optimised on the manipulated variables values of R_2 and B_2 (the two recirculations of the system).

The sequence have a double intermediate condition (D_1 and D_3 must be located in the boundary line). To solve the mass balances, is supposed that x_L^{D1} and x_H^{D1} must follow the boundary line equation. Then, x_L^{D3} and x_H^{D3} are calculated with the mass balances and then each valued are compared with the boundary line equation values (equation 22).

$$x_L^{D3} = x_{Lbl}^{D3} \qquad (22)$$

The same method as Setup 1 and Setup 2 is used to solve this problem and find the optimum values of R_2 and B_2.

Finally, the optimisation process is applied to all the range of crude feed compositions.

6. RESULTS

The results are visualized in a ternary Gibbs diagram for illustrative purposes. A critical comparison with Ulrich and Morari (2003) results is performed. The results are structured according to the different feasible Setups according to the ∞/∞ analysis.

6.1. SETUP 1

6.1.1. Mass balances for Setup 1

The mass balances that describe the system are available in section 5.3.1.3. Provided the feed stream and the two manipulated variables fulfilling the required degrees of freedom, then all compositions and streams of the sequence are calculated providing an insight about the relationships between the streams. The goal of the process is to separate the feed mixture in its compounds.

The output streams are able to reach pure compounds according to the ∞/∞ analysis assumptions. Hence output streams depend on the crude feed and the overall mass balances. R_2 (the stream flow rate recycled back) and D_3 (third column distillate flow rate) are used as input manipulated variables. Mass balances are presented in detail in Appendix 1.

6.1.2. Graphical solution of DSE respect of feed composition for Setup 1

The goal optimisation function requires the boiling temperatures at the top and bottoms of the columns because it depends on Carnot efficiency. The use of the simulator Aspen Plus® to get the boiling temperatures is unsuitable because compositions of the streams are changing with the range of feed composition. The information could be transferred between MATLAB® and Aspen Plus® or vice versa using Visual Basic®. However, the transfer of information between different software slow down very much the calculation procedure consuming more time the information transfer than calculations itself.

Simulis Thermodynamics® allows its direct implementation in Microsoft Excel ® as an Add-in or in MATLAB ® as a toolbox avoiding the need to transfer the results form one software to another. For this reason is used MATLAB ® with Simulis Thermodynamics ® wich allows to calculate fast and easily the boiling points of the mixtures for all the range of feed compositions and solve also in the same calculation environment the mass balances.

An iterative program covering all the feed compositions and optimizing R_2 and D_3 variables maximizing the DSE function is implemented in this work. Results of the DSE are represented in a 3D graphic in function of feed composition to identify when this Setup is more favourable depending on the feed composition (Figure 7) as Ulrich and Morari (2003).

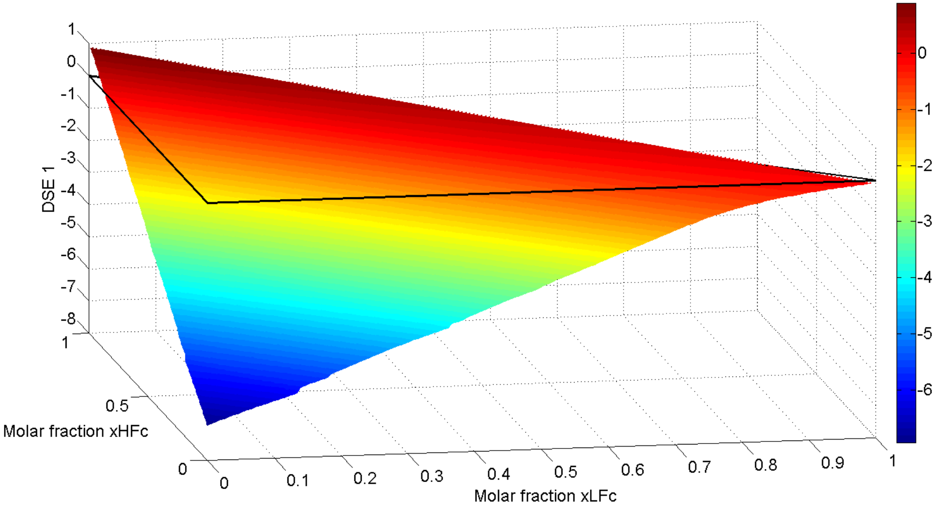


Figure 11: Graphical representation of DSE in front of crude feed composition for Setup 1

For Setup 1, DSE parameter is positive when composition of the light compound or of the heavy compound is high but is unfavourable and negative when the composition of the two compounds is small (Figure 9), i.e. when feed is mostly intermediate compound. The first separated compound is the heavy compound (*H*) (Figure 7). When H is the major component in crude feed, most of *H* is collected efficiently at the bottom of the first column. R_2 and D_3 are recycled streams and its back mixing provides a negative efficiency (recirculation streams have negative efficiency). As observed in equation 15 when most of the feed flow rate is collected at the first column bottom, R_2 and D_3 are small and positive DSE are obtained, on the other hand, when crude feed is rich in light and intermediate compounds, R_2 and D_3 becomes high and DSE becomes negative.

The light compound is collected at the top of column 2, when the crude feed is mostly light compound ($x_L^{Fc} \rightarrow 1$) , most of the crude feed is collected at the top of column 2 and *H* and *I* compounds are collected at column 2 bottoms. When most of the crude feed is collected at the top of column 2, the reboiler duties of columns 1 and 2 becomes very high as reflected in DSE close to 0 but, as D_3 tends to 0, because the major part of the flow rate exits for column 2, do not make DSE negative.

6.1.3. Graphical solution of DSE respect of degrees of freedom for Setup 1

To determine the effect of R_2 and D_3, Figure 12 shows DSE in function of these two recycle streams chosen as variables.

DSE is maximum when $R_2 \rightarrow 0$ and $D_3 \rightarrow 0$ but, there is a region where $D_3 \rightarrow 0$ and R_2 is not 0 when DSE is positive. This region corresponds to the zone where the crude feed is mainly composed of light compound.

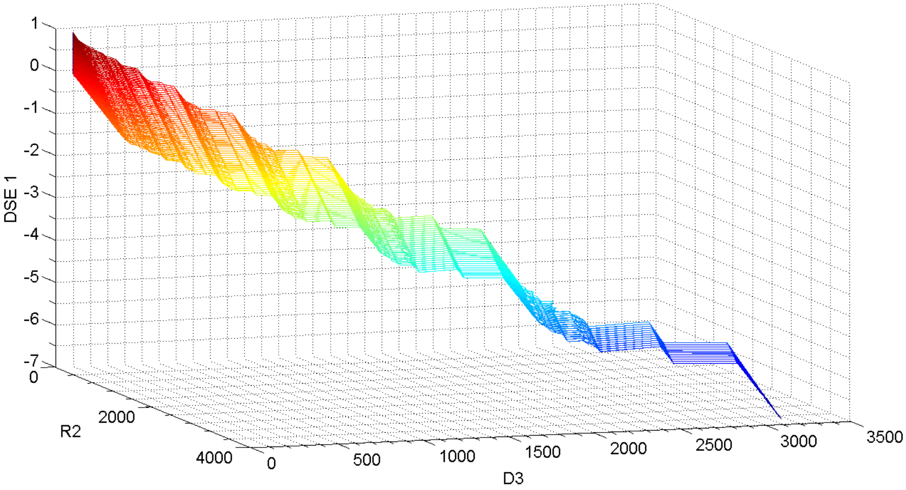


Figure 12: Graphical representation of DSE in front of degrees of freedom for Setup 1

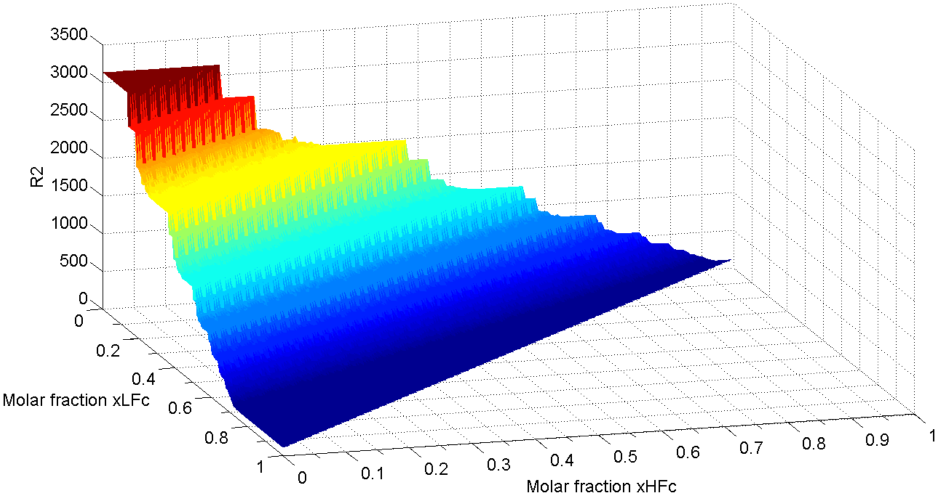


Figure 13: Graphical representation of R_2 in front of crude feed composition in Setup 1

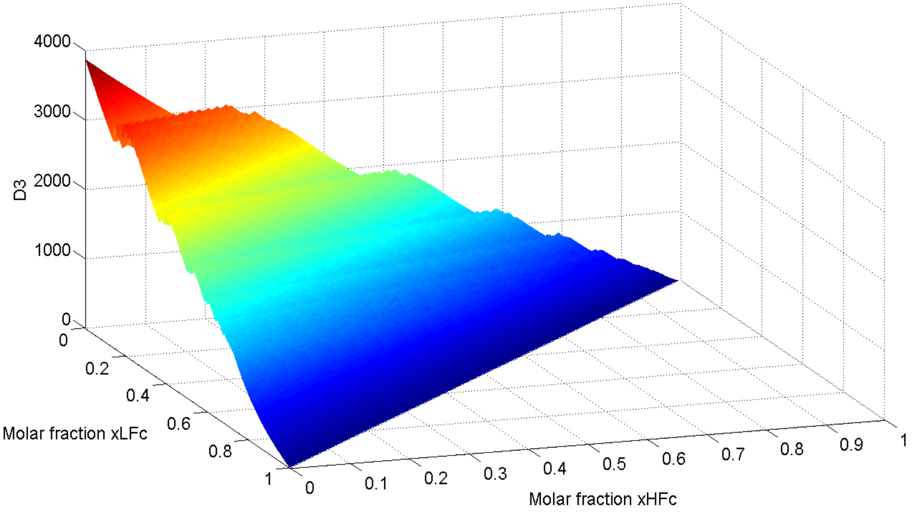


Figure 14: Graphical representation of D_3 in front of crude feed composition for Setup 1

Figures 13 and 14 shows optimal R_2 and D_3 depending on crude feed composition. DSE in Setup 1 is maximum in the absence of *I* compound and when R_2 and D_3 are minimum. In the absence of *L* or *H* compounds, R_2 and D_3 are maximum what generates negative DSEs. In the presence of high amounts of *L* or *H* compounds in crude feed, the recirculation streams tend to 0 and maximizing the DSE for this Setup.

6.2. SETUP 2

6.2.1. Mass balances for Setup 2

As in the case of Setup 1, mass balances must be performed to describe the sequence and find the relationships between al the streams. ∞/∞ analysis is used and the same streams are supposed because they are degrees of freedom. Mass balances equations are presented in Appendix 1.

6.2.2. Graphical solution of DSE respect of feed composition for Setup 2

As Setup 1, the optimisation function requires the boiling temperatures at the top and bottoms of the columns because it depends on Carnot efficiency. Hence another MATLAB® program is made with Simulis Thermodynamics® to create an iterative procedure which calculates the optimal values of the optimisation variables.

Results are visualized in a ternary Gibbs diagram as Setup 1 in function of feed composition as Ulrich and Morari (2003).

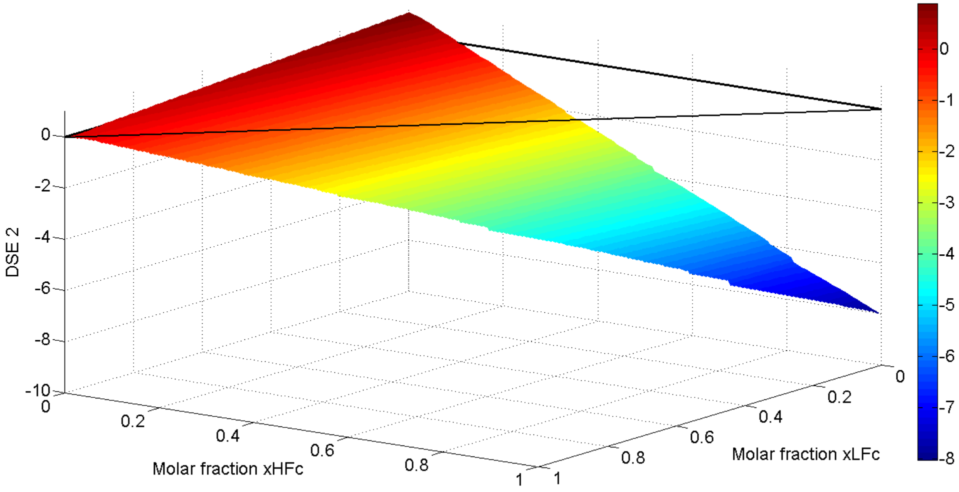


Figure 15: Graphical representation of DSE in front of crude feed composition for Setup 2

For Setup 2, DSE parameter is positive when the composition of heavy compound in the feed tends to 0 ($x_H^{Fc} \rightarrow 0$) (Figure 15). This is because in Setup 2, the light compound is the first to be separated in the first column which is easily to be separated (his efficiency would be very high). In the bottom of column 2 the intermediate compound is collected, if a high quantity of heavy compound is collected at the top of the column, this needs a lot of energy because it is not an easily separation and this makes that the efficiency decreases. In addition to this, if crude feed composition is mostly *L* compound, R_2 may be high hence in column 1 entry a lot of *L* compound that makes that D_1 have a very high flow rate. As illustrated in Figure 9, D_1 is the only recirculation stream that decreases the efficiency. This is reflected in Figure 15 where for a high composition of *L*, the efficiency is approximately 0.

When most of the feed is intermediate compound, the sequence presents the highest efficiency because *L* compound is collected easily in the first column and all of intermediate compound is collected at the bottom of column 2 while high quantity of *I* is collected efficiently at the bottom of column 3. At the top of column 2 a small quantity of *H* and *I* compounds is collected and mixed with a small quantity of *L* (R2), which generates a small value of flow rate at the top of the column (D_1) that does not affect DSE practically.

6.2.3. Graphical solution of DSE respect of degrees of freedom for Setup 2

To determinate the effect of R_2 and D_3 Figure 16 illustrates DSE in function of the two recycle streams chosen variables.

DSE is maximum when R_2 and D_3 are minimum (tend to 0). This As seen in Figure 16 and explained in section 6.2.2, the feed flow rate of column 1 is very high, the recirculation flow rate (D_1) is very high too, therefore DSE function as seen in equation 18 provides a small value because D_1 has negative efficiency.

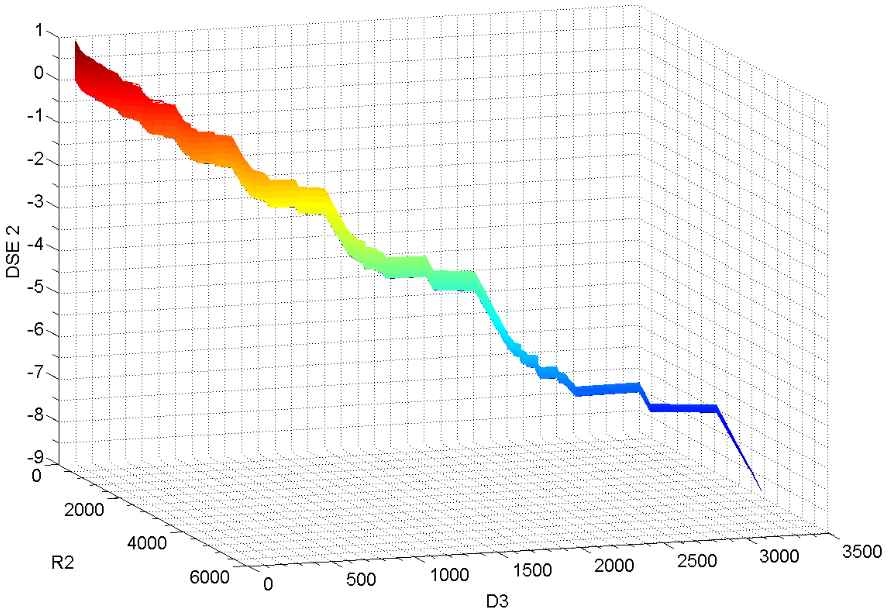


Figure 16: Graphical representation of DSE in front of degrees of freedom for Setup 2

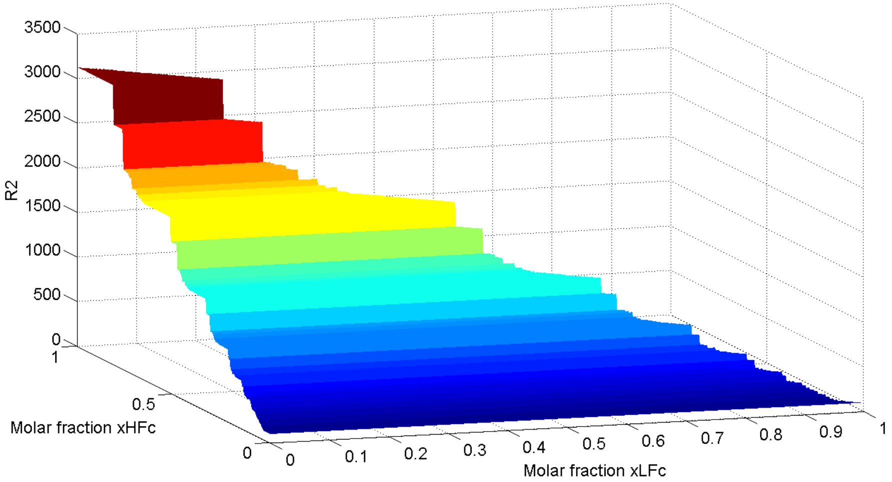


Figure 17: Graphical representation of R_2 in front of crude feed composition for Setup 2

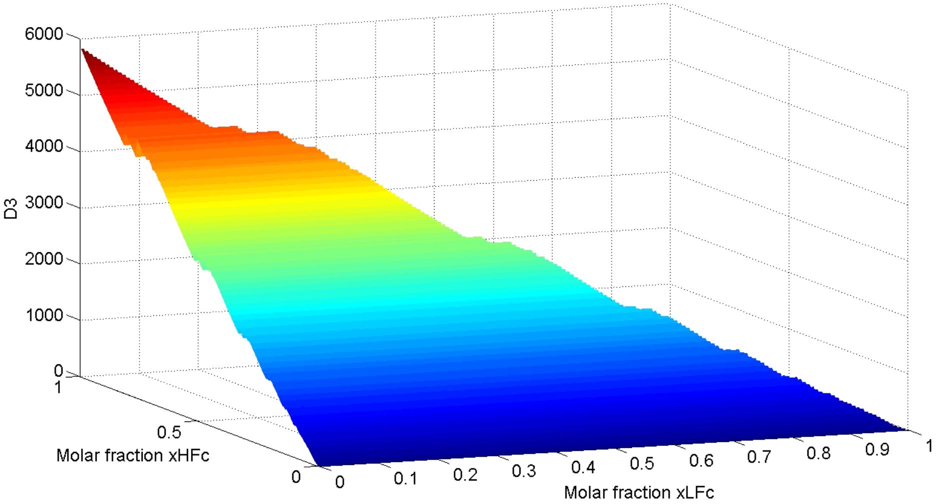


Figure 18: Graphical representation of D_3 in front of crude feed composition for Setup 2

Figures 17 and 18 illustrate optimal R_2 and D_3 depending on crude feed composition. These two parameters affect the value of the DSE function (if they are too high, DSE decreases). As these figures show, in the absence of *H* compound, R_2 and D_3 are minimal, hence DSE function would be very high.

6.3. SETUP 3

6.3.1. Mass balances for Setup 3

Mass balances are done to describe the system and know the relations between the streams. Output streams according to the ∞/∞ analysis assumptions are fixed by the crude feed (pure compounds). In this Setup, R_2 and B_2 are used as input variables because they are the variables chosen to fulfil the two required degrees of freedom. Mass balances are presented in detail in Appendix 1.

6.3.2. Graphical solution of DSE respect of feed composition for Setup 3

Another iterative program with MATLAB® using the boiling temperatures at the top and bottoms of the columns of Simulis Thermodynamics® is made to find the optimal results of the optimisation variables.

Results of the DSE are represented in a 3D graphic in function of feed composition as Ulrich and Morari (2003).

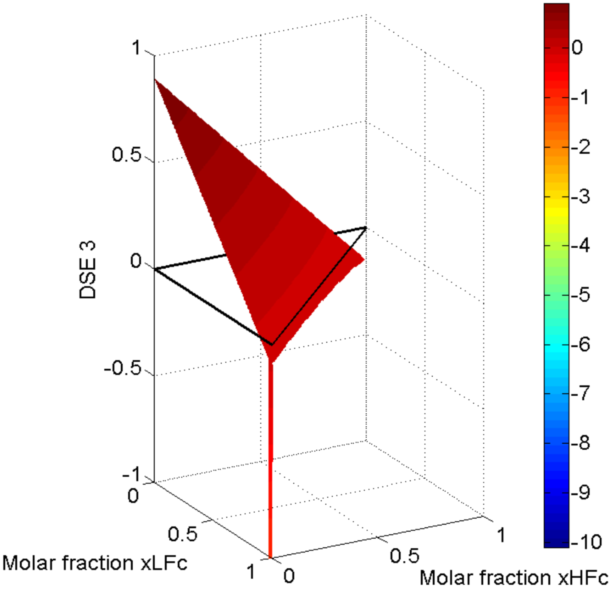


Figure 19: Graphical representation of DSE in front of crude feed composition for Setup 3

For Setup 3, DSE function is positive for the major range of compositions (Figure 19). The highest values of DSE are when the most part of feed stream is intermediate compound because in column 3 intermediate compound (*I*) is collected in the bottom with the maximum efficiency.

At the top of the column, a little part of *H* compound is collected with a little part of *I* and *L* compounds that do not need a lot of energy to be separated hence efficiency in this column is very high.

The other streams of the sequence are not very high hence recirculation streams with negative efficiency are very small and not affect the DSE function.

When feed composition is pure *L* compound, the DSE function decreases a lot (to negative values).This is because the *L* compound is collected at the top of all the columns wasting a lot of energy and decreasing DSE values.

6.3.3. Graphical solution of DSE respect degrees of freedom for Setup 3

Figure 20 illustrates the effect of R_2 and B_2 on the value of DSE.

To maximise DSE, R_2 must have a fixed value (102.2 kmol/h for a calculus base of 1,000 kmol/h) and B_2 (depends on the crude feed composition) changes its value. As seen in Figure 20, B_2 varies because when crude feed is mainly composed of *L* compound, the DSE function tends to negative values. To maximise DSE, B_2 tends to 0 to minimize the recycle stream causes the decrease of DSE function.

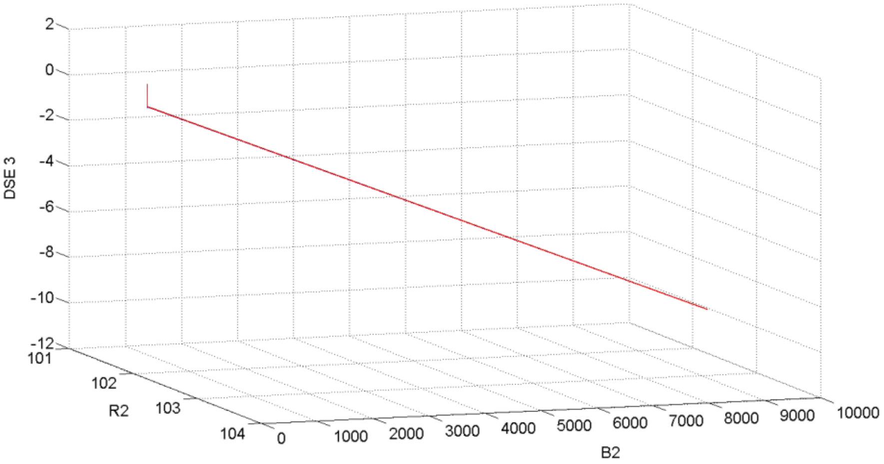


Figure 20: Graphical representation of DSE in front of degrees of freedom for Setup 3

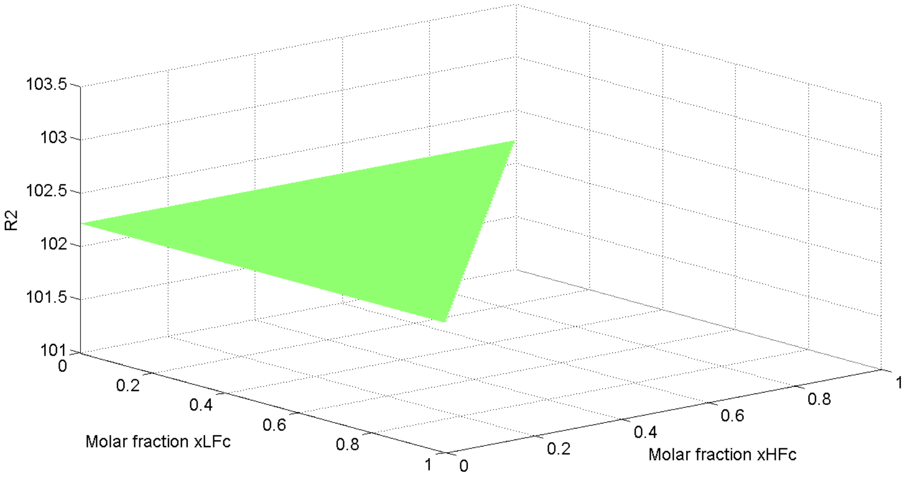


Figure 21: Graphical representation of R_2 in front of crude feed composition for Setup 3

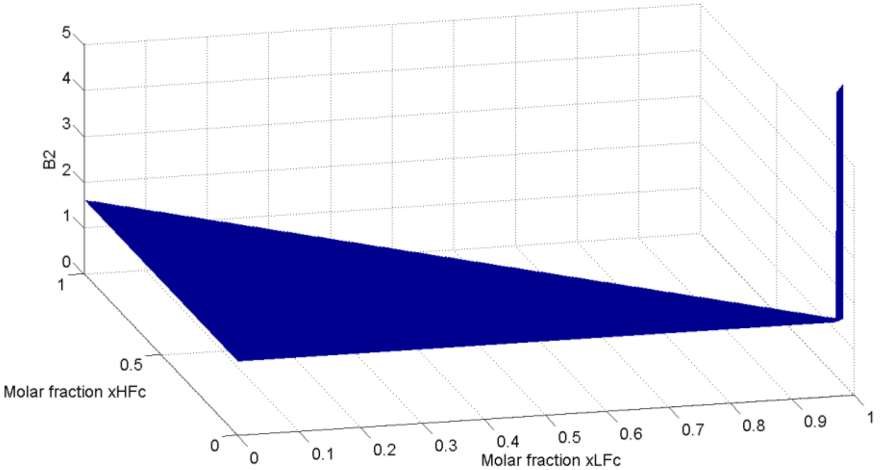


Figure 22: Graphical representation of B_2 in front of crude feed composition for Setup 3

R_2 does not depend on crude feed composition as can be seen in Figure 21 because always have the same value. B_2 only depends on feed composition if the main compound of the crude feed is the light compound (Figure 22). This makes DSE to tend to negative values and as (Figure 20) causing the variation of B_2 value. For any other composition, B_2 have a constant value approximately as to 1.5 kmol/h.

6.4. COMPARISON OF THE THREE SETUPS

In previous sections, the three Setups are commented separately; in this section, the three Setups are compared to identify the zones where DSE is better.

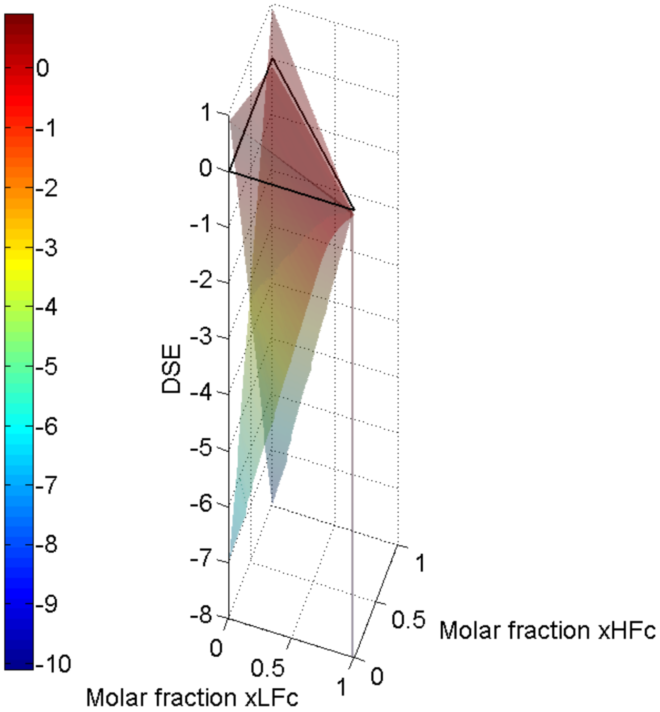


Figure 23: Graphical comparison of DSE function for the three Setups in 3-D

Figure 23 shows the optimal values for the three Setups at the optimal operating point for each crude feed composition. Setup 3 is the only sequence where for all the range of compositions have a positive DSE, except when the feed stream is mostly *L* compound. Hence, in general, Setup 3 is the best sequence to perform this separation but, as Figure 23 shows, Setup 3 is not the best in all the ranges of compositions, because for small quantities of *I* compound the other two Setups provide higher DSE values.

Figure 24 provides a more clear comparison of the three Setups, highlighting better the ranges of compositions in which each Setup performs best.

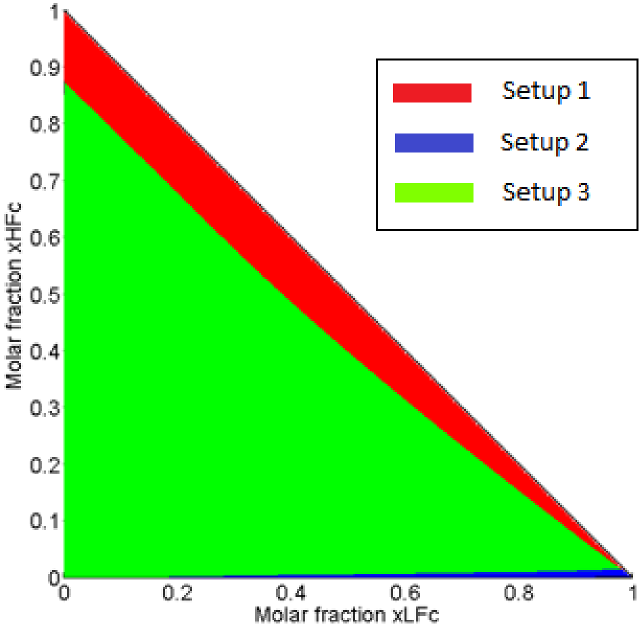


Figure 24: Graphical comparison of DSE function for the three setups in 2-D

Setup 1 is better for *L/H* mixtures or pure *H* compound feeds because the energy required for the separation is high when *I* compound appears in the mixtures. This is because the intermediate compound is collected at the top of all the distillation columns and in the third column, the separation of *I* and *H* compounds is difficult due to their close boiling points, decreasing the efficiency.

Setup 2 is better for binary *I/L* feeds or pure *L* for the same reason as in Setup 1 case. The heavy compound is collected at the top of all distillation columns and in the third column of the sequence, it must be separated from I compound decreasing the efficiency.

Setup 3 is better when crude feed is *I* compound pure or the three compounds are more or less in the same proportion because *I* and *H* compounds are separated first. Hence, the energy consumed to collect *L* compound at the top of all the columns is not very high.

As can be seen in Figures 21 and 22, B_2 and R_2 are fixed to a not so high value (R_2 = 102,2 kmol/h and B_2 = 1,2 kmol/h) with respect to the crude feed flow rate. Hence, recirculation streams do not affect the DSE values, considering the other parameters of the optimisation function.

6.5. COMPARISON WITH ULRICH AND MORARI (2003) OPTIMISATION FUNCTION

In this section result obtained in section 6.4 are compared to Ulrich and Morari (2003) results to show the performance of the proposed optimisation function and its influence on to the selection of the most optimum Setup function of crude feed composition.

Ulrich and Morari (2003) optimise the sequences to obtain the minimum cost in distillation process taking into account that operability cost depends on distillation streams (equation 4) but do not take into account if one mixture is more easily or more difficult to be separated.

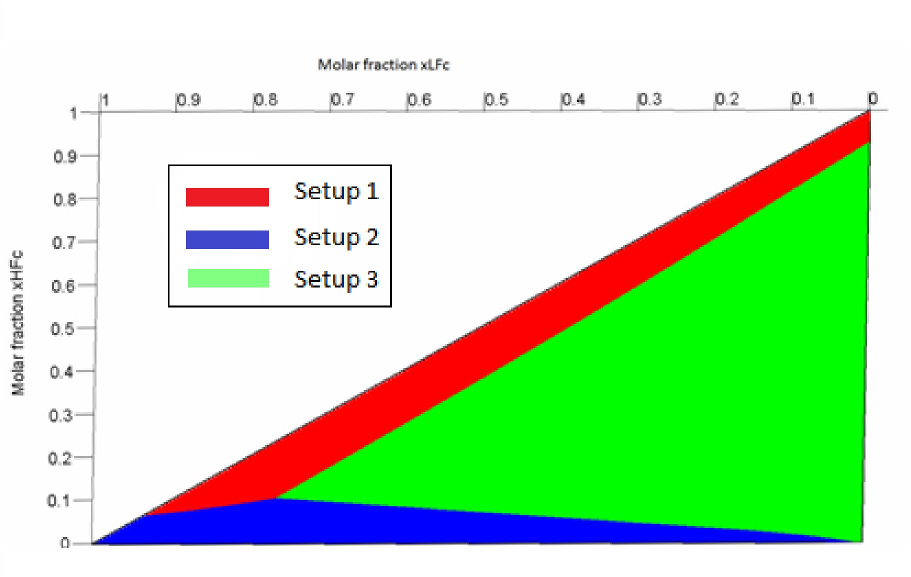


Figure 25: Graphical representation of Ulrich optimisation function for the three Setups (coloured zones provides the minimum operation cost)

Figure 25 shows that for a feed with a small quantity of *I* compound and an *L/H* mixture, Setup 1 provides the lowest operability cost because, as illustrated in Figure 7, *I* compound is collected at the top of columns 1 and 3, if the quantity of *I* compound is very high. In this case, D_1 and D_3 have high values as well, therefore the cost function value is consequently high. In case there is not *I* compound and considering an *L/H* mixture, all the distillation flow rates would not be so high.

For a high concentration of *L* compound, Setup 2 provides the lowest operability cost because, as seen in Figure 9, *L* compound as is collected at the top of column 2 with a high flow rate but if all the *L* compound is collected for D_2, D_1 and D_3 will be zero because all the *L* compound is collected previously.

Finally, if *I* is the main compound of the crude feed or if there is a mixture of the three compounds in the same proportion, Setup 3 provides the lowest cost function. As seen in Figure 10, if all the crude feed is intermediate compound, the major part of this stream is collected in the bottom of column 3 hence D_1, D_2 and D_3 will be 0 or approximately 0 so the cost is very small. If the crude feed proportion is more or less the same for all the compounds, D_1, D_2 and D_3 are not 0 but neither very high.

If we take into account the efficiency, the results obtained comparing Figures 24 and 25 similar because Figure 25 represents the streams D_1, D_2 and D_3 found in the DSE optimisation, for this reason the conclusion is the same.

6.6. COMPARISON WITH ULRICH AND MORARI (2003) RESULTS

When considering the original results of Ulrich and Morari (2003), Setup 2 provides the maximum DSE (being more efficient) for high *L* compound concentration. For high *I* concentration, Setup 2 is not the better sequence perform the separation, this result being different from the Ulrich and Morari (2003) results that state Setup 2 to be the best for high *I* contents.

For high or medium contents of *H*, as illustrated in Figure 4, Setup 1 provides the lowest cost function because when *H* is the major compound, for the first column *H* compound is collected at the bottom hence distillation streams of the three columns are very small. When efficiency is taken into account, for high *H* contents, Setup 1 is the best coinciding with Ulrich and Morari (2003) but for medium contents of *H*, Setup 3 is the more efficient Setup.

Finally, Setup 3 and Setup 2 in Ulrich and Morari (2003) studies provide more or less the same operation cost function for a ternary feed but Setup 3 gives the highest operation cost function. For a binary feed *I/H,* Setup 3 provides a lowest cost function because when feed entry directly in column 3 (*I* compound is separated at the bottom) and then in column 1 (*H* compound is separated at the bottom) the *I* and *H* compounds that are collected at the bottom of column 1 and column 3, do not pass by the reboiler of column 2, which decreases the cost function. But if we take into account the efficiency, Setup 3 is the best sequence for high *I* compound and for the maximum range of compositions as illustrated in Figure 24.

6.7 COMPARISON WITH HEURISTICS

For a simple distillation sequence, heuristics are commonly used to find an efficient separation method. For an azeotropic distillation sequence with recycled streams, heuristics are not easily applicable.

Figure 24 illustrate that the heuristic rule that says that the first compound to separate is the main compound is accomplished because in Setup 1, the highest efficiency is when crude feed stream is pure *H* compound which is the first compound to be separated.

In Setup 2 and Setup 3 happens the same, the highest efficiencies are when crude feed stream is formed by *L* and *I* pure compounds respectively. Therefore, the results using the DSE method are in agreement with the heuristics.

The heuristics are qualitative and the DSE method allows quantifying at which concentrations the "main compound" can be considered. The heuristic of favouring the direct sequence is also fulfilled due to the fact that Setup 3 covers the widest concentration region. The heuristics to perform the easiest/cheapest separation first are not always fulfilled because the presence of a easily separable compound could act as extracting agent or entrainer helping to perform difficult separations.

10. CONCLUSIONS

The conclusions are:

- The operation of a sequence of three homogeneous azeotropic distillation columns with two recycles (boundary separation scheme) separating a three-component mixture into pure components is analyzed.

- A program is implemented done to find the best Setup for this mixture but, it can be adapted easily to any other mixture.

- The analysis of the system using simplified models provides a solution for this distillation sequence.

- Studying a case with reduced complexity (columns of infinite length operated at infinite reflux) showed that taking into account the efficiency, Setup 3 gives the highest range of compositions that gives the best DSE.

- The results are compared changing the optimisation function. The results where similar when the efficiency is taken into account.

- The results are compared with previous studies taking only the required energy. Ulrich and Morari (2003) claim that Setup 3 should not be used, but when the efficiency is taken into account, Setup 3 has the greatest range of application.

11. REFERENCES AND NOTES

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APPENDICES

APPENDIX 1: MASS BALANCES

SETUP 1

First Assumptions

$$Fc = 1000\,\frac{kmol}{h}$$

$$x_L^{Fc} = 0{,}30$$

$$x_H^{Fc} = 0{,}30$$

$$x_I^{Fc} = 0{,}40$$

$$x_H^{B1} = 1$$

$$x_L^{D2} = x_L^{R2} = x_L^{D2ex} = 1$$

$$x_I^{B3} = 1$$

$$x_H^{D3} = x_I^{D3} = 0{,}5$$

$$x_L^{D1} = 2(x_H^{D1})^2 - 3x_H^{D1} + 1$$

$$R_2 = degree\ of\ freedom$$

$$D_3 = dregree\ of\ freedom$$

H compound mass balance in all the system

$$0 = x_H^{Fc} \cdot Fc - x_H^{B1} \cdot B_1 \rightarrow B_1 = \frac{x_H^{Fc} \cdot Fc}{x_H^{B1}}$$

I compound mass balance in all the system

$$0 = x_I^{Fc} \cdot Fc - x_I^{B3} \cdot B_3 \rightarrow B_3 = \frac{x_I^{Fc} \cdot Fc}{x_I^{B3}}$$

Overall mass balance in all the system

$$0 = Fc - B_1 - D_2^{ex} - B_3 \rightarrow D_2^{ex} = Fc - B_1 - B_3$$

Overall mass balance in branch knot $\mathbf{D_2 \rightarrow R_2 + D_2^{ex}}$

$$0 = D_2 - R_2 + D_2^{ex} \rightarrow D_2 = R_2 + D_2^{ex}$$

Overall mass balance in column number 3

$$0 = B_2 - D_3 - B_3 \rightarrow B_2 = D_3 + B_3$$

H compound mass balance in column number 3

$$0 = x_H^{B2} \cdot B_2 - x_H^{D3} \cdot D_3 \rightarrow x_H^{B2} = \frac{x_H^{D3} \cdot D_3}{B_2}$$

$$x_I^{B2} = 1 - x_H^{B2}$$

Overall mass balance in branch knot $\mathbf{R_2 + D_3 \rightarrow M_1}$

$$0 = R_2 + D_3 - M_1 \rightarrow M_1 = R_2 + D_3$$

L compound mass balance in branch knot $\mathbf{R_2 + D_3 \rightarrow M_1}$

$$0 = x_L^{M1} \cdot M_1 - x_L^{R2} \cdot R_2 \rightarrow x_L^{M1} = \frac{x_L^{R2} \cdot R_2}{M_1}$$

H compound mass balance in branch knot $\mathbf{R_2 + D_3 \rightarrow M_1}$

$$0 = x_H^{M1} \cdot M_1 - x_H^{D3} \cdot R_2 \rightarrow x_H^{M1} = \frac{x_H^{D3} \cdot R_2}{M_1}$$

$$x_I^{M1} = 1 - x_L^{M1} - x_H^{M1}$$

Overall mass balance in branch knot $\mathbf{Fc + M_1 \rightarrow F_1}$

$$0 = Fc + M_1 - F_1 \rightarrow F_1 = Fc + M_1$$

L compound mass balance in branch knot $\mathbf{Fc + M_1 \rightarrow F_1}$

$$0 = x_L^{M1} \cdot M_1 + x_L^{Fc} \cdot Fc - x_L^{F1} \cdot F_1 \rightarrow x_L^{F1} = \frac{x_L^{M1} \cdot M_1 + x_L^{Fc} \cdot Fc}{F_1}$$

H compound mass balance in branch knot $\mathbf{Fc + M_1 \rightarrow F_1}$

$$0 = x_H^{M1} \cdot M_1 + x_H^{Fc} \cdot Fc - x_H^{F1} \cdot F_1 \rightarrow x_H^{F1} = \frac{x_H^{M1} \cdot M_1 + x_H^{Fc} \cdot Fc}{F_1}$$

$$x_I^{F1} = 1 - x_L^{F1} - x_H^{F1}$$

Overall mass balance in column number 1

$$0 = F_1 - D_1 - B_1 \rightarrow D_1 = F_1 - B_1$$

L compound mass balance in column number 1

$$0 = x_L^{F1} \cdot F_1 - x_L^{D1} \cdot D_1 \rightarrow x_L^{D1} = \frac{x_L^{F1} \cdot F_1}{D_1}$$

H compound mass balance in column number 1

$$0 = x_H^{F1} \cdot F_1 - x_H^{D1} \cdot D_1 - x_H^{B1} \cdot B_1 \rightarrow x_H^{D1} = \frac{x_H^{F1} \cdot F_1 - x_H^{B1} \cdot B_1}{D_1}$$

$$x_I^{D1} = 1 - x_L^{D1} - x_H^{D1}$$

SETUP 2

First assumptions

$Fc = 1000\frac{kmol}{h}$ $\quad x_H^{B1} = 1$ $\quad x_L^{D1} = 2(x_H^{D1})^2 - 3x_H^{D1} + 1$

$x_L^{Fc} = 0{,}30$ $\quad x_L^{D2} = x_L^{R2} = x_L^{D2ex} = 1$ $\quad R_2 = degree\ of\ freedom$

$x_H^{Fc} = 0{,}30$ $\quad x_I^{B3} = 1$ $\quad D_3 = dregree\ of\ freedom$

$x_I^{Fc} = 0{,}40$ $\quad x_H^{D3} = x_I^{D3} = 0{,}5$

L compound mass balance in all the system

$$0 = x_L^{Fc} \cdot Fc - x_L^{D2ex} \cdot D_2^{ex} \rightarrow D_2^{ex} = \frac{x_L^{Fc} \cdot Fc}{x_L^{D2ex}}$$

I compound mass balance in all the system

$$0 = x_I^{Fc} \cdot Fc - x_I^{B3} \cdot B_3 \rightarrow B_3 = \frac{x_I^{Fc} \cdot Fc}{x_I^{B3}}$$

Overall mass balance in all the system

$$0 = Fc - D_2^{ex} - B_3 - B_1 \rightarrow B_1 = Fc - D_2^{ex} - B_3$$

Overall mass balance in branch knot $\mathbf{D_2 \rightarrow R_2 + D_2^{ex}}$

$$0 = D_2 - R_2 + D_2^{ex} \rightarrow D_2 = R_2 + D_2^{ex}$$

Overall mass balance in branch knot $\mathbf{R_2 + D_3 \rightarrow M_1}$

$$0 = R_2 + D_3 - M_1 \rightarrow M_1 = R_2 + D_3$$

L compound mass balance in branch knot $\mathbf{R_2 + D_3 \rightarrow M_1}$

$$0 = x_L^{M1} \cdot M_1 - x_L^{R2} \cdot R_2 \rightarrow x_L^{M1} = \frac{x_L^{R2} \cdot R_2}{M_1}$$

H compound mass balance in branch knot $\mathbf{R_2 + D_3 \rightarrow M_1}$

$$0 = x_H^{M1} \cdot M_1 - x_H^{D3} \cdot R_2 \rightarrow x_H^{M1} = \frac{x_H^{D3} \cdot R_2}{M_1}$$

$$x_I^{M1} = 1 - x_L^{M1} - x_H^{M1}$$

Overall mass balance in column number 3

$$0 = B_2 - D_3 - B_3 \rightarrow B_2 = D_3 + B_3$$

H compound mass balance in column number 3

$$0 = x_H^{B2} \cdot B_2 - x_H^{D3} \cdot D_3 \rightarrow x_H^{B2} = \frac{x_H^{D3} \cdot D_3}{B_2}$$

$$x_I^{B2} = 1 - x_H^{B2}$$

Overall mass balance in column number 1

$$0 = M_1 - B_1 - D_1 \rightarrow D_1 = M_1 - B_1$$

L compound mass balance in column number 1

$$0 = x_L^{M1} \cdot M_1 - x_L^{D1} \cdot D_1 \rightarrow x_L^{D1} = \frac{x_L^{M1} \cdot M_1}{D_1}$$

I compound mass balance in column number 1

$$0 = x_I^{M1} \cdot M_1 - x_I^{D1} \cdot D_1 \rightarrow x_I^{D1} = \frac{x_I^{M1} \cdot M_1}{D_1}$$

$$x_H^{D1} = 1 - x_L^{D1} - x_I^{D1}$$

Overall mass balance in branch knot $\boldsymbol{D_1 + Fc \rightarrow F_2}$

$$0 = D_1 + Fc - F_2 \rightarrow F_2 = D_1 + Fc$$

L compound mass balance in branch knot $\boldsymbol{D_1 + Fc \rightarrow F_2}$

$$0 = x_L^{D1} \cdot D1 + x_L^{Fc} \cdot Fc - x_L^{F2} \cdot F_2 \rightarrow x_L^{F2} = \frac{x_L^{D1} \cdot D1 + x_L^{Fc} \cdot Fc}{F_2}$$

H compound mass balance in branch knot $\boldsymbol{D_1 + Fc \rightarrow F_2}$

$$0 = x_H^{D1} \cdot D_1 + x_H^{Fc} \cdot Fc - x_H^{F2} \cdot F_2 \rightarrow x_H^{F2} = \frac{x_H^{D1} \cdot D_1 + x_H^{Fc} \cdot Fc}{F_2}$$

$$x_I^{D1} = 1 - x_L^{F2} - x_H^{F2}$$

SETUP 3

First assumptions

$Fc = 1000\frac{kmol}{h}$ $x_H^{B1} = 1$ $x_L^{D1} = 2(x_H^{D1})^2 - 3x_H^{D1} + 1$

$x_L^{Fc} = 0{,}30$ $x_L^{D2} = x_L^{R2} = x_L^{D2ex} = 1$ $x_L^{D3} = 2(x_H^{D3})^2 - 3x_H^{D3} + 1$

$x_H^{Fc} = 0{,}30$ $x_I^{B3} = 1$ $R_2 = degree\ of\ freedom$

$x_I^{Fc} = 0{,}40$ $B_2 = degree\ of\ freedom$

H compound mass balance in all the system

$$0 = x_H^{Fc} \cdot Fc - x_H^{B1} \cdot B_1 \rightarrow B_1 = \frac{x_H^{Fc} \cdot Fc}{x_H^{B1}}$$

L compound mass balance in all the system

$$0 = x_L^{Fc} \cdot Fc - x_L^{D2ex} \cdot D_2^{ex} \rightarrow D_2^{ex} = \frac{x_L^{Fc} \cdot Fc}{x_L^{D2ex}}$$

Overall mass balance in all the system

$$0 = Fc - B_3 - B_1 - D_2^{ex} \rightarrow B_3 = Fc - B_1 - D_2^{ex}$$

Overall mass balance in branch knot $\boldsymbol{R_2 + D_2^{ex} \rightarrow D_2}$

$$0 = R_2 + D_2^{ex} - D_2 \rightarrow D_2 = R_2 + D_2^{ex}$$

Overall mass balance in column number 2

$$0 = D_1 - D_2 - B_2 \rightarrow D_1 = D_2 + B_2$$

L compound mass balance in column number 2

$$0 = x_L^{D1} \cdot D_1 - x_L^{D2} \cdot D_2 \rightarrow x_L^{D1} = \frac{x_L^{D2} \cdot D_2}{D_1}$$

D1 must stay in the boundary line

$$0 = 2(x_H^{D3})^2 - 3x_H^{D3} + 1 - x_L^{D1} \rightarrow x_H^{D1} = Iteration$$

H compound mass balance in column number 2

$$0 = x_H^{D1} \cdot D_1 - x_H^{B2} \cdot B_2 \rightarrow x_H^{B2} = \frac{x_H^{D1} \cdot D_1}{B_2}$$

Overall mass balance in branch knot $\boldsymbol{Fc + B_2 \rightarrow F_3}$

$$0 = Fc + B_2 - F_3 \rightarrow F_3 = Fc + B_2$$

L compound mass balance in branch knot $\boldsymbol{Fc + B_2 \rightarrow F_3}$

$$0 = x_L^{Fc} \cdot Fc - x_L^{F3} \cdot F_3 \rightarrow x_L^{F3} = \frac{x_L^{Fc} \cdot Fc}{F_3}$$

H compound mass balance in branch knot $\boldsymbol{Fc + B_2 \rightarrow F_3}$

$$0 = x_H^{Fc} \cdot Fc - x_H^{B2} \cdot B_2 \rightarrow x_H^{Fc} = \frac{x_H^{B2} \cdot B_2}{Fc}$$

$$x_I^{F3} = 1 - x_L^{F3} - x_H^{F3}$$

Overall mass balance in column number 3

$$0 = F_3 - D_3 - B_3 \rightarrow D_3 = F_3 - B_3$$

L compound mass balance in column number 3

$$0 = x_L^{F3} \cdot F_3 - x_L^{D3} \cdot D_3 \rightarrow x_L^{D3} = \frac{x_L^{F3} \cdot F_3}{D_3}$$

H compound mass balance in column number 3

$$0 = x_H^{F3} \cdot F_3 - x_H^{D3} \cdot D_3 \rightarrow x_H^{D3} = \frac{x_H^{F3} \cdot F_3}{D_3}$$

$$x_I^{D3} = 1 - x_L^{D3} - x_H^{D3}$$

Overall mass balance in branch knot $\boldsymbol{D_3 + R_2 \rightarrow M_1}$

$$0 = D_3 + R_2 - M_1 \rightarrow M_1 = D_3 + R_2$$

H compound mass balance in branch knot $\boldsymbol{D_3 + R_2 \rightarrow M_1}$

$$0 = x_H^{D3} \cdot D_3 - x_H^{M1} \cdot M_1 \rightarrow x_H^{M1} = \frac{x_H^{D3} \cdot D_3}{M_1}$$

I compound mass balance in branch knot $\boldsymbol{D_3 + R_2 \rightarrow M_1}$

$$0 = x_I^{D3} \cdot D_3 - x_I^{M1} \cdot M_1 \rightarrow x_I^{M1} = \frac{x_I^{D3} \cdot D_3}{M_1}$$

$$x_L^{M1} = 1 - x_H^{M1} - x_I^{M1}$$