

Design of non-equilibrium stage separation systems by a stochastic optimization approach for a class of mixtures



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ABSTRACT

Design and analysis of distillation columns usually takes place by using rigorous equilibrium models, where efficiencies are utilized to enhance the designs obtained and have a better approach to the real performance of the column. A second alternative, based on the mass and heat transfer rates, has been proposed and it is known as the non-equilibrium (or rate-based) model. By employing this approach, the use of supposed values for efficiencies is not necessary. Nevertheless, to the authors' knowledge, there is not a short-cut design model for analysis of distillation columns with the non-equilibrium model. In this work, the design and optimization of conventional and intensified distillation sequences, with equilibrium and non-equilibrium models for the separation of ternary mixtures, is presented. The multiobjective optimization is performed by a stochastic technique with handling constraints, which is also coupled to a process simulator. This strategy allows analyzing the distillation systems with the complete rigorous models (MESH and MERSHQ equations, respectively). Optimal designs are then compared to determinate relationships between the design parameters of the optimal equilibrium sequences and the optimal non-equilibrium sequences. It has been found that the optimal on heat duty for each analyzed systems is almost the same for both, equilibrium and non-equilibrium models, but with different number of stages. Nevertheless, other design variables have only slight differences between models for an optimal structure.

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1. Introduction

Distillation is one of the most used separation processes on chemical and petrochemical industries. It is a well-known process and its design and control has been studied since many years ago. Nevertheless, its main drawback is its inherent low second-law efficiency, which results into high external energy requirements. To enhance the energetic performance of distillation for ternary mixtures, the use of thermally coupled distillation systems has been proposed and analyzed; it has been showed that those schemes may be helpful to obtain energy savings of about 30% when compared to the conventional distillation sequences [1–7].

For difficult separations, the use of hybrid processes as extractive distillation has been proved as a good alternative for achieving high purities of the products. Such systems may have lower energy requirements and lower total annual costs than the conventional trains [8–10]. Design and analysis of distillation systems is usually performed by using the so-called equilibrium model, which considers that phase equilibrium is reached on each stage of the column. Nevertheless, equilibrium is rarely achieved on a real tray, thus, efficiencies are used to take into account deviations from the ideal performance of the tray. Since efficiencies are not known a priori, their values are supposed when designing a distillation column, and usually a single, constant value for efficiency is considered for all the trays in the column. This assumption may lead to under or over-designing the column. Thus, an alternative model was proposed by Taylor and Krishna, which considers the mass and heat transfer rates on each tray and assumes equilibrium

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Nomenclature

Q_i (kJ/h)	heat duty of the column i
Q_T (kJ/h)	total heat duty
N_i	number of stages of the column i
N_T	total number of stages
N_{EQ}	number of stages for the equilibrium model
N_{NEQ}	number of stages for the non-equilibrium model
F_{solv} (kmol/h)	molar flow rate of solvent
\mathbf{y}_m	vector of obtained purities for the m components
\mathbf{x}_m	vector of required purities for the m components

only on the interface [11] this model is known as non-equilibrium (NEQ) model. The successful use of the non-equilibrium model for the analysis of intensified sequences has been reported previously [12–15].

Agreement of the results obtained by simulations with the non-equilibrium model and experimental data has also been reported [15,16]. Different design and optimization methodologies have been proposed, such as parametric analysis [12,13], orthogonal collocation [17], simulated annealing [18,19] and differential evolution [20]. The aforementioned non-equilibrium optimization approaches consider just one objective, usually the total annual cost. In this work, a multiobjective genetic algorithm, with constraints handling, coupled to the process simulator Aspen Plus is used for the optimization of conventional and intensified distillation systems by using the equilibrium and the non-equilibrium model. The genetic algorithm is accelerated with artificial neural networks, as proposed by Gutiérrez-Antonio and Briones-Ramírez [21]. Furthermore, the genetic algorithm is linked to the process simulator Aspen Plus, thus the rigorous models of the systems are already available; and it can be used for the optimization. Optimal designs for both models are compared in terms of Pareto fronts, in order to obtain guidelines for the design of low-cost non-equilibrium distillation systems taking as a basis the equilibrium designs.

2. Case of study

In order to cover a representative range of analyzed mixtures, different components and compositions have been considered; with thermodynamic behavior ranging from ideal to non-ideal.

Furthermore, ordinary and intensified distillation sequences have been studied. The distillation systems presented in this work can be grouped as follows:

- Ordinary distillation sequences (conventional and thermally coupled).
- Extractive distillation (conventional and thermally coupled).

The mixtures considered for separation in ordinary distillation sequences are shown in Table 1, together with the feed composition, recovery and desired purity. The first mixture consists of alcohols: methanol (MEOH), ethanol (ETOH) and butanol (BUOH); with two feed compositions: low and high composition of the middle-boiling component. This mixture will be separated on a conventional direct sequence (Fig. 1a) and a thermally coupled direct sequence (Fig. 1b). In Fig. 1, A, B and C represent the light, middle and heavy components, respectively. The second mixture consists of hydrocarbons with the presence of an isomer: isopentane (IC5), n -pentane (NC5) and n -hexane (NC6), with two different feed compositions. This mixture will be treated on a conventional indirect sequence (Fig. 1c) and a thermally coupled indirect sequence (Fig. 1d).

To consider mixtures far from ideality, systems with the presence of azeotropes have been studied. Those mixtures have been purified using extractive distillation to achieve high purities of the components. In Table 2, azeotropic systems analyzed in this work are presented. The first azeotropic mixture has acetone (ACET) and methanol (MEOH), for which a minimum boiling azeotrope has been reported at a composition of 77.6 mol% of acetone [22]. This mixture is separated in an extractive distillation direct sequence (both conventional and thermally coupled, Fig. 2a and b, respectively) using water as solvent to obtain purities higher than the azeotropic ones. The second azeotropic mixture consists on ethanol (ETOH) and water (H_2O), where a maximum boiling azeotrope exists between ethanol and water for purities higher than 90 mol% of ethanol (about 96 wt%) [23,24]. This mixture is also purified in an extractive direct sequence, using ethylene glycol as solvent. The trays for all the distillation systems (conventional, extractive and thermally coupled) are Sieve trays.

The stochastic optimization strategy used in this work is explained in Section 3.

3. Optimization tool

A multiobjective genetic algorithm with constraints handling [21] has been used for the optimization of the proposed distillation sequences. The genetic algorithm is coupled to the process simulator Aspen Plus, thus the whole equilibrium and

Table 1
Mixtures analyzed for ordinary distillation.

Mixture	Components	Feed composition (mol%)	Recovery (mol%)	Desired purity (mol%)
M1F1	Methanol	0.4	0.99	0.995
	Ethanol	0.2	0.98	0.961
	Butanol	0.4	0.99	0.995
M1F2	Methanol	0.15	0.99	0.955
	Ethanol	0.7	0.98	0.996
	Butanol	0.15	0.99	0.955
M2F1	Isopentane	0.4	0.99	0.995
	n -Pentane	0.2	0.98	0.961
	n -Hexane	0.4	0.99	0.995
M2F2	Isopentane	0.15	0.99	0.955
	n -Pentane	0.7	0.98	0.995
	n -Hexane	0.15	0.99	0.955

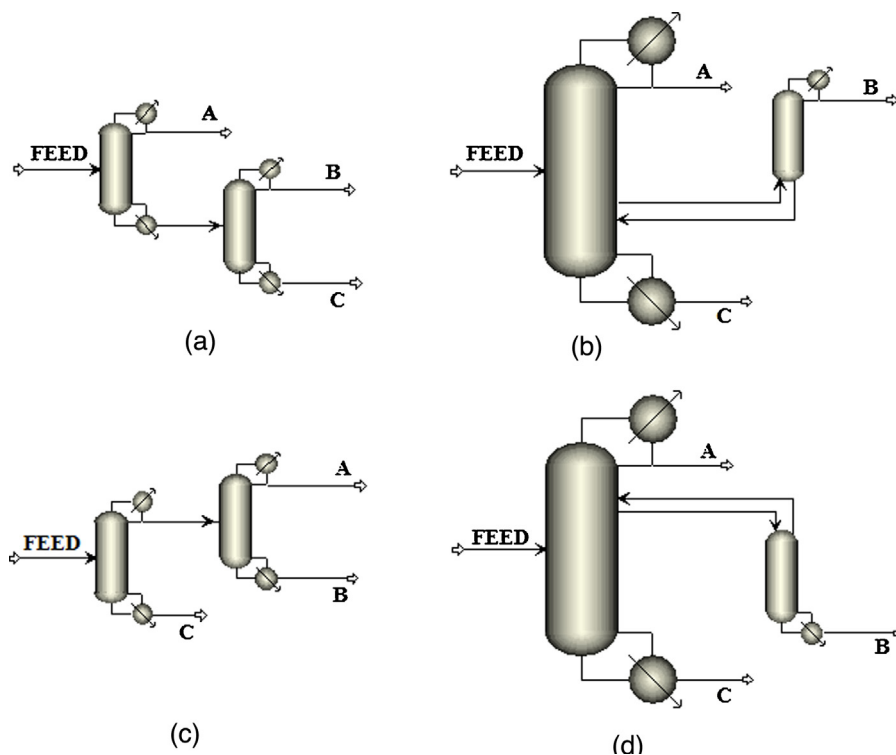


Fig. 1. Ordinary distillation sequences (a) direct conventional sequence, (b) thermally coupled direct sequence (TCDS), (c) indirect conventional sequence, and (d) thermally coupled indirect sequence (TCIS).

Table 2
Mixtures analyzed for distillation with azeotropes.

Mixture	Components	Feed composition (mol%)	Recovery (mol%)	Desired purity (mol%)
M3F1	Acetone	0.5	0.998	0.995
	Methanol	0.5	0.998	0.995
	Water (S)	1.0	0.998	0.9995
M4F2	Ethanol	0.1	0.999	0.999
	Water	0.9	0.994	0.99
	Ethylene glycol (S)	1.0	0.982	0.99

non-equilibrium models are respectively used for the optimization of the distillation systems. The algorithm generates a set of optimal designs that represent the best trade-off between the selected objectives; it manages several objectives simultaneously, manipulating integer and continue variables, and the link to Aspen Plus allows considering the complete model of the distillation columns (MESH and MERSHQ equations). The genetic

algorithm is based on NSGA-II [25], and the constraints are handling using a multiobjective technique [26]; due to this, penalty functions are avoided in the handling of the constraints, while the use of weights is not necessary for the objectives. The code is implemented in Matlab, which is linked to Aspen Plus using ActiveX Technology. More details about the algorithm can be found in the original work [21].

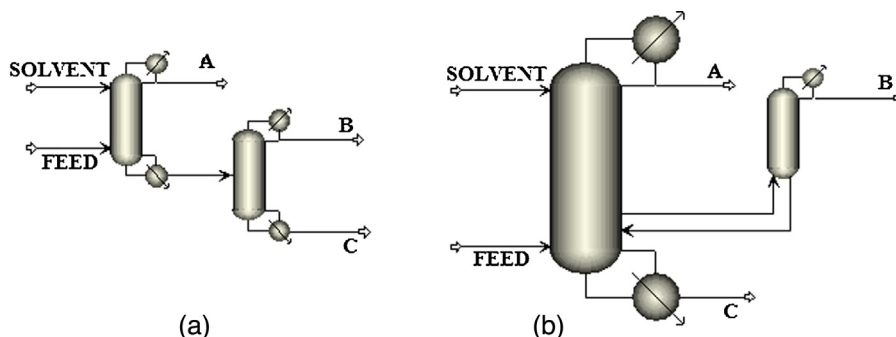


Fig. 2. Extractive distillation sequences (a) direct conventional extractive sequence and (b) thermally coupled extractive direct sequence (TCEDS).

Table 3

Objectives to be simultaneously minimized to generate the Pareto fronts of ordinary distillation systems.

	Direct conventional	Indirect conventional	Direct coupled	Indirect coupled
Number of stages in column 1, N_1	X	X	X	X
Number of stages in column 2, N_2	X	X	X	X
Heat duty in column 1, Q_1	X	X	X	X
Heat duty in column 2, Q_2	X	X	X	X
Total number of objectives	4	4	3	4

The algorithm works as follows: the first generation is created from an initial design, and all individuals are sent to Aspen Plus to simulate them and get data about objectives and constraints; then, individuals are ranked in subpopulations, according to the number of satisfied constraints. The best individuals are selected to be the new parents of the second generation, and so on; inside each subpopulation, the fulfilment of the constraints is considered as another objective to optimize. Finally, the process ends when the maximum number of generations has been reached. One of the more slow steps in the optimization process is the evaluation of the objective function; thus, the algorithm is modified with neuronal networks to enhance the speed of the evaluations. As result of the optimization process the Pareto Front of the distillation sequences is obtained. The Pareto front is integrated by all optimal designs that represent the best trade-offs between the objectives of interest.

The optimization problem for the ordinary distillation sequences, both conventional and thermally coupled, involves the simultaneous minimization of the number of stages and the heat duty in each column of the sequence; this minimization problem is subject to the required recoveries and purities in each product stream:

$$\text{Min}(Q_i, N_i) \text{ subject to}$$

$$\mathbf{y}_m \geq \mathbf{x}_m \quad (1)$$

where Q_i and N_i are the heat duty and the number of stages of the column i , while \mathbf{y}_m and \mathbf{x}_m are vectors of obtained and required purities for the m components, respectively. The particular number of objectives for both conventional and thermally coupled distillation sequences is presented in Table 3, for equilibrium

and non-equilibrium models. This minimization implies the manipulation of several variables as continuous as integer with the equilibrium and non-equilibrium model, respectively, which are presented in Table 4. Note that since the product streams flows are manipulated, the recoveries of the key components in each product stream must be included as a restriction, which are presented in Tables 1 and 2 for each study case. It can be noticed that the diameter of the columns is a manipulable variable when the non-equilibrium model is used. This is because the diameter of the trays affects the heat and mass transfer, thus it has a direct impact on energy requirements. Nevertheless, the diameter of the column cannot be freely manipulated because of the hydraulic limitations, also included in the model and, of course, in the optimization strategy.

The optimization problem for the extractive distillation sequences, both conventional and thermally coupled, involves the simultaneous minimization of the number of stages and the heat duty in each column of the sequence along with the flow of solvent; this minimization problem is subject to the required recoveries and purities in each product stream:

$$\text{Min}(Q_i, N_i, F_{\text{solv}}) \text{ subject to}$$

$$\mathbf{y}_m \geq \mathbf{x}_m \quad (2)$$

where Q_i and N_i are the heat duty and the number of stages of the column i and F_{solv} is the flow of solvent required for extraction, while \mathbf{y}_m and \mathbf{x}_m are vectors of obtained and required purities for the m components, respectively. The particular number of objectives for both conventional and thermally coupled extractive distillation sequences is presented in Table 5, for equilibrium and non-equilibrium models. This minimization implies the

Table 4

Manipulated variables to generate the Pareto fronts of each ordinary distillation systems.

	Direct conventional	Indirect conventional	Direct coupled	Indirect coupled
Equilibrium model				
Reflux ratio in column 1, R_1	X	X	X	X
Feed stage in column 1, $N_{F,1}$	X	X	X	X
Number of stages in column 1, N_1	X	X	X	X
Reflux ratio in column 2, R_2	X	X		
Feed stage in column 2, $N_{F,2}$	X	X		
Number of stages in column 2, N_2	X	X	X	X
Distillate stream flow of column 1	D1		D	D
Distillate stream flow of column 2	D2	D2	S	
Bottoms stream flow of column 2		B2		S
Product stage of liquid interconnection flow FL in column i , $N_{FL,i}$				$N_{FL,1,C1}$
Feed stage of vapor interconnection flow FV in column i , $N_{FV,i}$				$N_{FV,1,C1}$
Vapor interconnection flow, FV			FV1	
Feed stage of liquid interconnection flow FL in column i , $N_{FL,i}$			$N_{FL,1,C1}$	
Product stage of vapor interconnection flow FV in column i , $N_{FV,i}$			$N_{FV,1,C1}$	
Liquid interconnection flow, FL				FL1
Total	8	8	9	9
Non-equilibrium model (additional variables considered)				
Diameter of column 1, Diam_1	X	X	X	X
Diameter of column 2, Diam_2	X	X	X	X
Total	10	10	11	11

Table 5

Objectives to be simultaneously minimized to generate the Pareto fronts of extractive distillation systems.

	Direct conventional	Indirect conventional	Direct thermally coupled	Indirect thermally coupled
Number of stages in column 1, N_1	X	X	X	X
Number of stages in column 2, N_2	X	X	X	X
Heat duty in column C1, Q_1	X	X	X	X
Heat duty in column C2, Q_2	X	X	X	X
Flow of solvent, F_{solv}	X	X	X	X
Total number of objectives	5	5	4	5

manipulation of several variables as continuous as integer with the equilibrium and non-equilibrium model, respectively, which are presented in Table 6. Note that since the product streams flows are manipulated, the recoveries of the key components in each product stream must be included as a restriction, which are presented in Tables 1 and 2 for each study case. In order to determine the number of generations and the number of individuals required a tuning procedure is used. For all systems 2000 individuals have been used. The numbers of generations were 120, 160, 200 and 240 for conventional, thermally coupled, extractive conventional and extractive thermally coupled systems, respectively.

4. Methodology

Process simulator Aspen Plus has been considered for the analysis of the distillation sequences. The general strategy followed to the optimization of the distillation sequences is:

1. Generation of an initial design in equilibrium which allows obtaining the desired purities and recoveries.
2. Generation of an initial design in non-equilibrium, taking as a basis the designs obtained in the first step.
3. Optimization of the equilibrium and non-equilibrium designs by coupling the process simulator Aspen Plus with a genetic algorithm decoded in MATLAB, using the parameters described in Section 3.
4. Analysis of the obtained Pareto front of distillation sequences with equilibrium and non-equilibrium models.

The particular methodology for obtaining initial designs for each kind of distillation sequences will be presented next.

4.1. Ordinary distillation sequences

For the mixtures M1F1, M1F2, M2F1 and M2F2, a feed molar flow rate of 100 kmol/h has been considered. In the case of the mixture of alcohols, vapor–liquid equilibrium (VLE) has been calculated by using the NRTL model. Residual curves for the mixture of alcohols are shown in Fig. 3a. In the case of the hydrocarbon mixture, Chao–Seader model is considered as appropriate for these calculations. In Fig. 3b, residual curves for the system with isomers are shown. From ternary diagrams, it can be seen that both mixtures have an ideal performance, thus, conventional distillation can be used to separate their components. All feed streams have been considered as saturated liquid at 1.3 atm, which is the mean pressure between the top and the bottom of the columns. In order to obtain an initial guess for the designs of the conventional sequences, the Winn–Underwood–Gilliland method (DSTW module in Aspen Plus) has been used. With this information, the sequences have been simulated with the equilibrium model option selected in the RadFrac. In this stage, short-cut designs are tested to verify they allow obtaining the desired purities and recoveries. Then, taking as a basis the equilibrium designs, non-equilibrium simulations take place using the RadFrac module with the “Rate-Based” option selected.

In order to obtain designs for the thermally coupled distillation sequences, the stage rearrangement methodology [2] is used, taking as a basis the corresponding conventional designs either in

Table 6

Manipulated variables to generate the Pareto fronts of each extractive distillation systems.

	Direct conventional	Indirect conventional	Direct coupled	Indirect coupled
Equilibrium model				
Reflux ratio in column 1, R_1	X	X	X	X
Feed stage in column 1, $N_{F,1}$	X	X	X	X
Feed stage of solvent flow in column 1, $N_{\text{Solv},1}$	X	X	X	X
Solvent stream flow fed in column 1, F_{solv}	X	X	X	X
Number of stages in column 1, N_1	X	X	X	X
Reflux ratio in column 2, R_2	X	X		
Feed stage in column 2, $N_{F,2}$	X	X		
Number of stages in column 2, N_2	X	X	X	X
Distillate stream flow of column 1	D1		D	D
Distillate stream flow of column 2	D2	D2	S	
Bottoms stream flow of column 2		B2		S
Product stage of liquid interconnection flow FL in column i , $N_{\text{FL},i}$				$N_{\text{FL},C1}$
Feed stage of vapor interconnection flow FV in column i , $N_{\text{FV},i}$				$N_{\text{FV},C1}$
Vapor interconnection flow, FV			FV1	
Feed stage of liquid interconnection flow FL in column i , $N_{\text{FL},i}$			$N_{\text{FL},C1}$	
Product stage of vapor interconnection flow FV in column i , $N_{\text{FV},i}$			$N_{\text{FV},C1}$	
Liquid interconnection flow, FL				FL1
Total	10	10	11	11
Non-equilibrium model (additional variables considered)				
Diameter of column 1, Diam_1	X	X	X	X
Diameter of column 2, Diam_2	X	X	X	X
Total	12	12	13	13

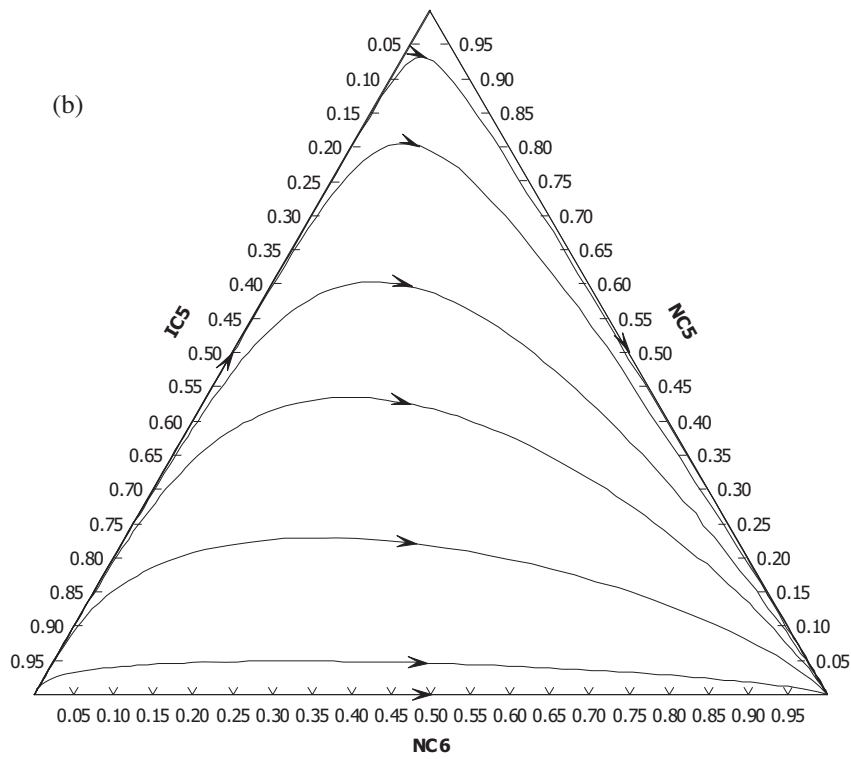
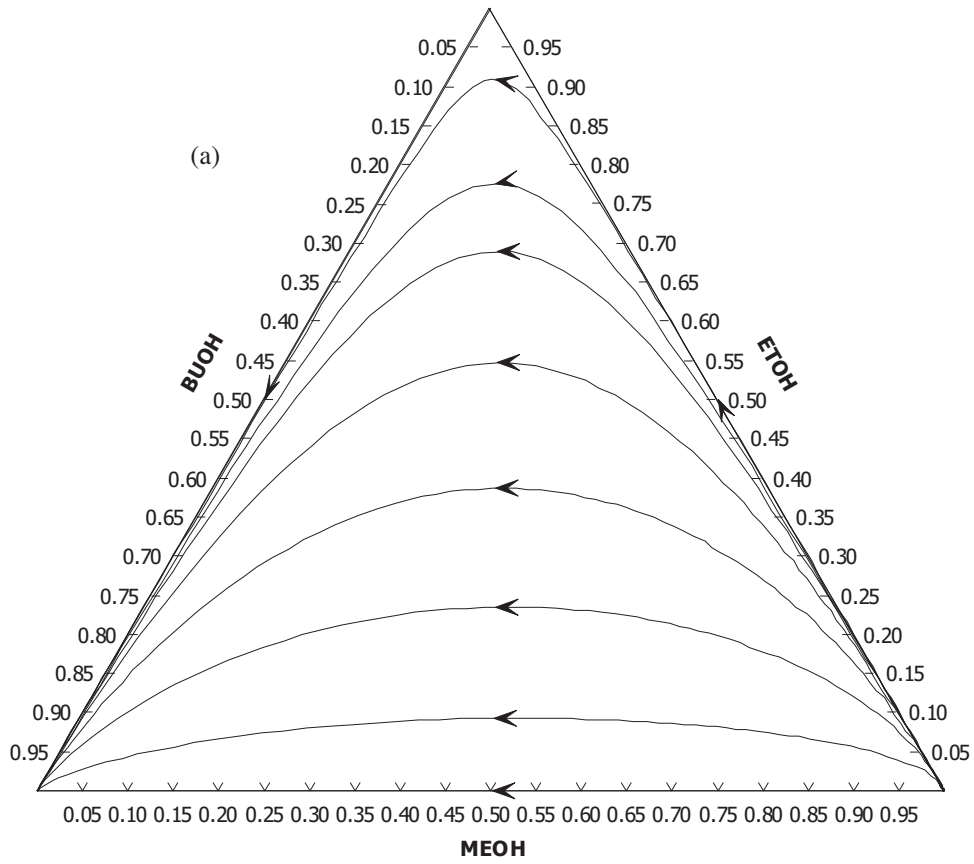


Fig. 3. Molar residual curves for conventional distillation systems (a) ethanol/*n*-butanol/methanol and (b) *n*-pentane/*i*-pentane/*n*-hexane.

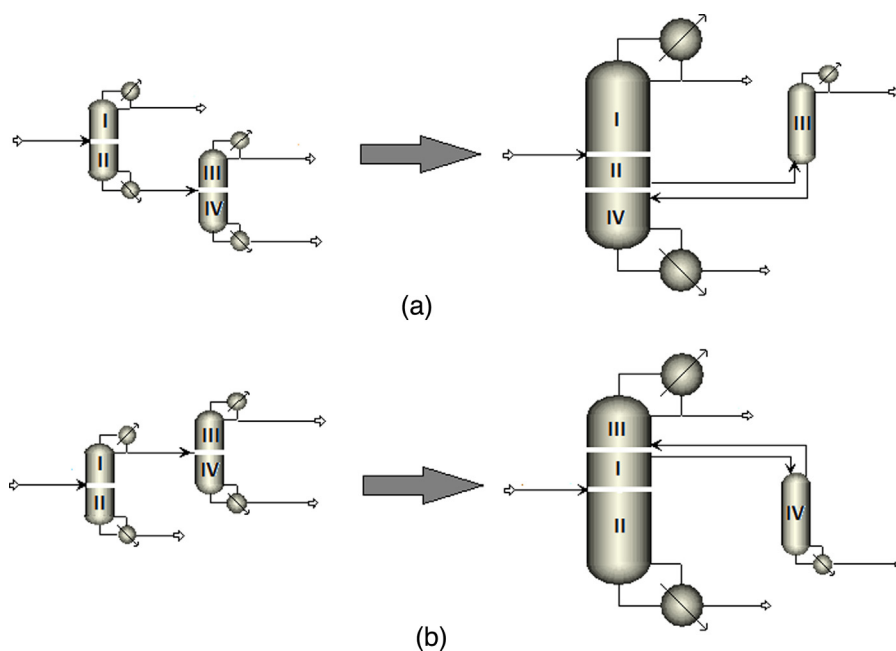


Fig. 4. Rearrangement of stages (a) from direct conventional sequence to TCDS and (b) from indirect conventional sequence to TCIS.

equilibrium or non-equilibrium. The stage rearrangement method will be briefly explained. From the short-cut methods, the number of stages and feed stage can be estimated for conventional sequences, for example the conventional direct sequence (Fig. 4a). To obtain the design of the thermally coupled direct sequence, the stages below the feed stage of the second column (section IV) are moved to the lower section of the first column. Thus, the number of stages in the main column is computed as the sum of the number of stages on sections I, II and IV. A similar procedure is used for the thermally coupled indirect sequence, as can be observed in Fig. 4b.

4.2. Extractive distillation sequences

In the case of the mixture M3F1, a feed molar flow rate of 540 kmol/h has been considered, since this molar flow rate has been studied by Luyben [27]. In order to model the vapor–liquid equilibrium for this mixture, the UNIQUAC model has been used [28]. It has been reported that the UNIQUAC model fits with vapor–liquid equilibrium experimental data for the system acetone/methanol/water [29]. In Fig. 5a, the ternary residual curves diagram is shown. The presence of an azeotropic point can be observed for the binary system acetone/ethanol at a temperature of 328.39 K. For mixture M4F2, a feed molar flow rate of 100 kmol/h has been considered. Thermodynamic model NRTL-RK has been used to analyze this mixture [23,24]. In Fig. 5b, the residual curves diagram for mixture M4F1 is shown. An azeotrope can be observed between water and ethanol at 351.29 K. The presence of azeotropes indicate non-idealities, this is why the use of an entrainer is necessary to achieve high purities.

Since there is not a short-cut method for designing extractive distillation columns, initial equilibrium design for conventional sequences have been obtained by assuming an initial number of stages, an initial feed flow rate for solvent and locating the solvent feed at the stage 2, while the feed of mixture to separate is fed at the stage N-1. Then, it has been verified if such configuration allows obtaining the desired purities and recoveries. Then, taking as a basis the equilibrium designs, the non-equilibrium systems are developed. Once the conventional systems accomplish the established purities and recoveries, the thermally coupled system

are designed by a stage rearrangement strategy, similar to the one used for ordinary distillation.

5. Results and discussion

5.1. Ordinary distillation sequences

From the multiobjective optimization procedure of each sequence, a Pareto front is obtained. The Pareto front is a set of optimal designs which represent the best compromises between the two objectives (number of stages and total heat duty). Thus, the Pareto front includes the designs with the lowest heat duty, the lowest number of stages, and various designs between those two extreme cases. In Fig. 6, the Pareto fronts obtained for the separation of mixture 1 are shown; we have grouped all the objective of interest in order to clarify the visualization of the results. In Table 7, the design parameters of the designs with the lowest and highest heat duty are shown with comparative purposes for the conventional systems. The diameter for the equilibrium simulations has been calculated by using the “Tray Sizing” option in Aspen Plus. It must be noticed that the stages are counted from top to bottom; thus stage 1 corresponds to the condenser of conventional columns. In Fig. 6, it can be seen that for the conventional direct sequence, heat duty is very similar for the optimal designs in equilibrium and non-equilibrium, but the non-equilibrium sequences have a higher number of stages. This occurs in a similar way for thermally coupled systems. For both models, thermally coupled sequences show lower energy requirements than the conventional sequences. It can be observed in Table 7 that the design with the minimum heat duty predicted by the non-equilibrium model requires about 1.5 times the number of equilibrium stages on the first column, and about 1.6 times the number of equilibrium stages for the second column. Nevertheless, other design variables, such as reflux, heat duty and diameter have very similar values for both models. If the whole Pareto front is analyzed, required non-equilibrium stages are about 1.4–2.6 times the number of equilibrium stages for the first column and about 1.4–3.1 times the equilibrium stages for the second column; it is worth to mention that the Pareto front is integrated by optimal

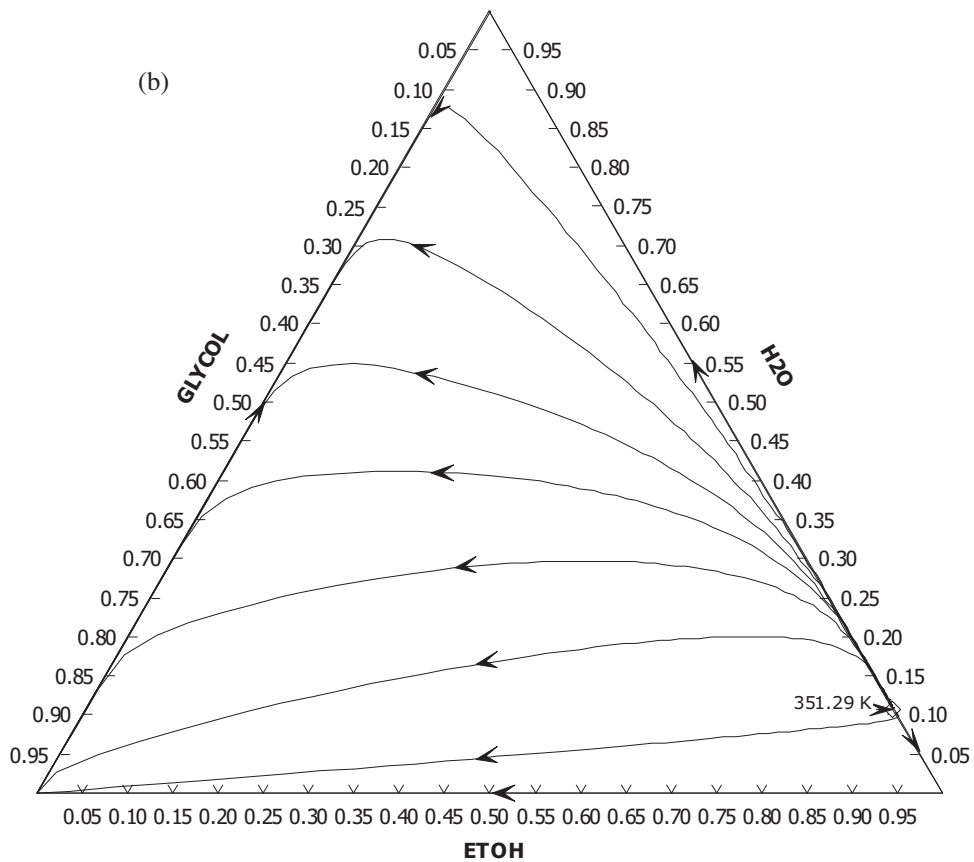
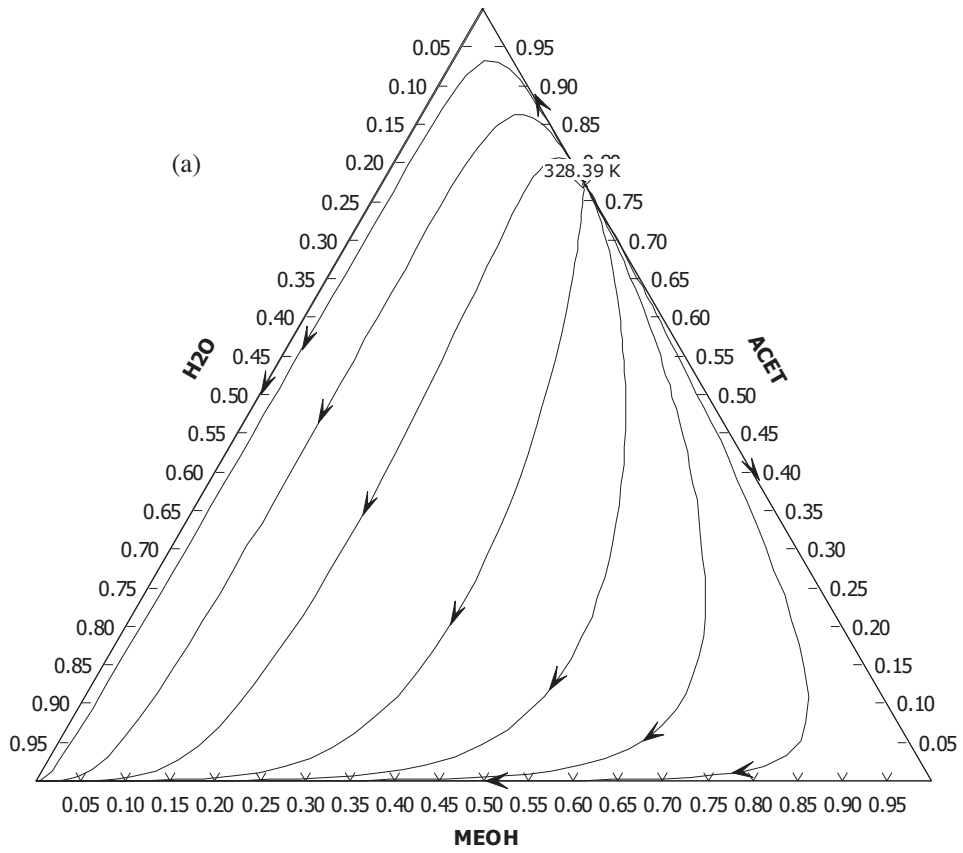


Fig. 5. Molar residual curves for extractive distillation systems (a) acetone/methanol/water and (b) ethanol/water/ethylene glycol.

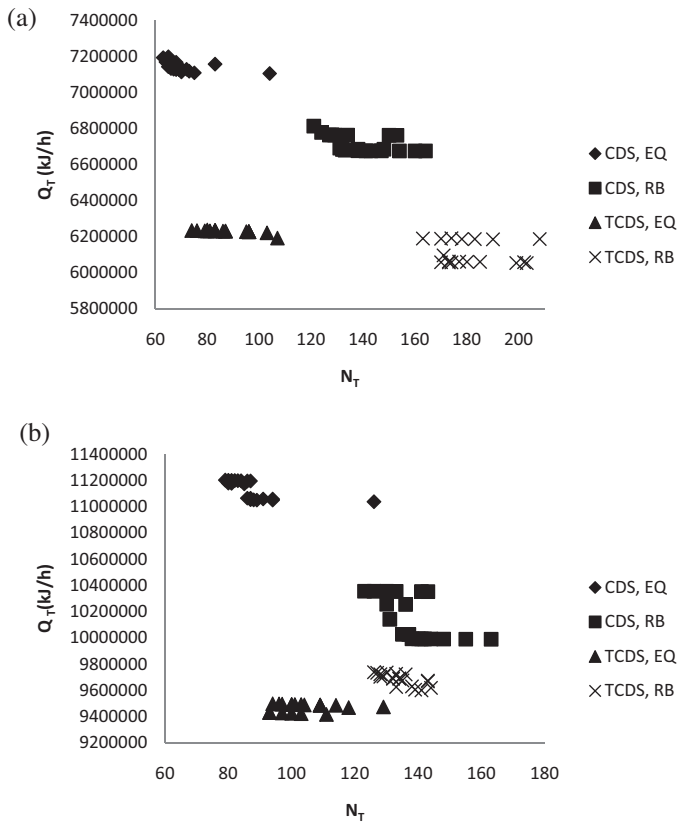


Fig. 6. Pareto front for the separation of mixture M1 in ordinary distillation sequences (a) F1 and (b) F2.

designs from minimum heat duty to minimum number of stages, and all designs between these extremes. Similar results have been obtained for the other mixtures. A summary of the ratios between the number of non-equilibrium and equilibrium stages for all cases is shown in Table 8 for the minimum heat duty case and for the whole Pareto front. It can be seen that for the conventional distillation sequences there is a wide range of N_{NEQ}/N_{EQ} ratios, which allows designs with low energy requirements. Nevertheless, the design with the minimum heat duty requires ratios close to the lower bound for the ratios on the whole Pareto front. It can also be noticed that the ratios required for the isomers mixture are higher than those for the alcohols mixture.

In Table 9, the design parameters for the thermally coupled systems are shown for the separation of mixture M1F1. Ratios

Table 7
Design parameters for selected cases of conventional distillation, M1F1.

	Q low		Q high	
	CDS, EQ	CDS, NEQ	CDS, EQ	CDS, NEQ
First column				
Number of stages	76	118	45	92
Feed stage	26	60	26	60
Reflux ratio	2.69	2.36	2.72	2.42
Diameter (m)	0.83	0.97	0.83	0.97
Second column				
Number of stages	28	46	20	29
Feed stage	12	14	9	14
Reflux ratio	1.03	1.08	1.11	1.15
Diameter (m)	0.5	0.68	0.51	0.65
Total heat duty (MJ/h)	7105.9	6676.8	7199.6	6814.5

Table 8
Summary of the results for the distillation sequences.

	N_{NEQ}/N_{EQ}			
	Q low, C1	Q low, C2	Pareto front, C1	Pareto front, C2
Conventional distillation				
M1F2	1.3	1.3	1.3–1.6	1.3–2.9
M2F1	1.7	1.6	1.7–3.4	1.3–1.8
M2F2	2.1	2.0	2.0–3.3	2.0–2.4
Thermally coupled distillation				
M1F1	2.25	0.53	1.8–3.0	0.5–1.3
M1F2	1.14	2.33	0.9–1.6	0.8–3.2
M2F1	1.56	1.41	0.9–1.6	0.8–2.0
M2F2	1.90	0.65	0.7–2.3	0.4–0.8
Extractive conventional distillation sequences				
M3F1	1.13	1.22	0.85–1.25	0.9–2.2
M4F2	1.64	0.91	1.00–1.93	0.41–2.4
Extractive thermally coupled distillation sequences				
M3F1	1.13	0.63	1.01–1.35	0.57–0.86
M4F2	1.68	2.2	1.55–1.75	0.46–2.2

between the number of stages in non-equilibrium and equilibrium for all the analyzed thermally coupled systems are shown in Table 8. In Table 8, it is shown that the thermally coupled designs predicted by the non-equilibrium model requires 0.7–3.0 times the number of equilibrium stages on the first column, and about 0.4–3.2 times the number of equilibrium stages for the second column. Particularly, a smaller number of non-equilibrium stages for the side column is required for the design with the lowest energy requirements for mixtures M1F1 and M2F2, while the ratio N_{NEQ}/N_{EQ} for those two cases is higher than for mixtures M1F2 and M2F1. As for the conventional cases, reflux ratio and heat duty have very similar values for both models, together with the interlinking flow rate. Diameters are only slightly higher for the non-equilibrium model.

5.2. Extractive distillation sequences

Pareto fronts for the separation of mixture M3F1 and M4F2 in extractive distillation sequences are shown in Fig. 7; we have grouped all the objective of interest in order to clarify the visualization of the results. In Table 10, design parameters of the sequences with the lowest and highest heat duty are presented for the separation of mixture M3F1 in conventional extractive systems. As occurred with the ordinary distillation sequences, for extractive distillation (either conventional or thermally coupled systems) heat duty results similar for the corresponding sequence in both equilibrium and non-equilibrium, with the

Table 9
Design parameters for selected cases of thermally coupled distillation, M1F1.

	Q low		Q high	
	TCDS, EQ	TCDS, NEQ	TCDS, EQ	TCDS, NEQ
First column				
Number of stages	90	203	68	169
Feed stage	38	87	32	84
Reflux ratio	2.39	2.28	2.43	2.28
Diameter (m)	0.88	1.05	0.87	1.05
Interlinking stages	50, 61	123, 129	48, 57	123, 129
Interlinking flow rate (kmol/h)	27.44	28.45	27.34	31.83
Second column				
Number of stages	17	9	12	5
Feed stage	17	9	12	5
Diameter (m)	0.38	0.56	0.38	0.57
Total heat duty (MJ/h)	6193.3	6053.9	6237.6	6192.8

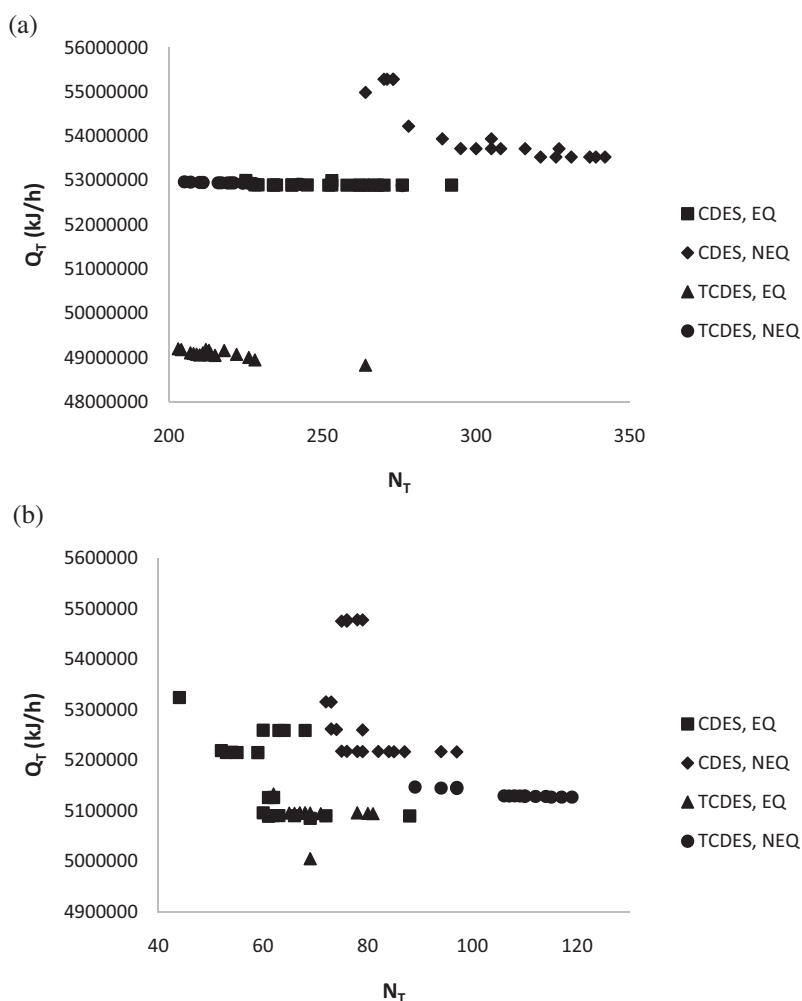


Fig. 7. Pareto front for the extractive ordinary distillation sequences (a) M3F1 and (b) M4F2.

non-equilibrium model predicting a higher number of stages. For mixture M3F1, direct thermally coupled extractive sequence shows lower energy requirements than the conventional extractive sequence. Similar results are obtained for mixture M4F2.

It can be observed on Table 8 that the conventional extractive design with the minimum heat duty predicted by the non-equilibrium model requires about 1.13 times the number of

equilibrium stages on the first column, and about 1.22 times the number of equilibrium stages for the second column. Reflux ratio and heat duty have very similar values for both models, while diameter is only slightly higher for the non-equilibrium design. If the whole Pareto front is analyzed, required non-equilibrium stages are about 0.85–1.25 times the number of equilibrium stages for the first column and about 0.9–2.2 times the equilibrium stages for the second column. For mixture M4F1, N_{NEQ}/N_{EQ} ratios from 1.00 to 1.93 for the first column and 0.41–2.4 for the second column allow obtaining the desired purities with low energy requirements.

In Table 11, the design parameters for the thermally coupled extractive systems are shown for the separation of mixture M3F1. Ratios between the number of stages in non-equilibrium and equilibrium for all the analyzed thermally coupled systems are shown in Table 8. From Table 8, it can be observed that for mixture M3F1 the extractive thermally coupled designs predicted by the non-equilibrium model requires 1.01–1.35 times the number of equilibrium stages on the first column, and about 0.57–0.86 times the number of equilibrium stages for the second column. For mixture M4F2 the ratios are 1.55–1.75 for the first column and 0.46–2.2 for the second column. It can be seen that, for the separation of mixture M3F1 with the lowest heat duty, the first column requires a similar number of stages in equilibrium and non-equilibrium. Nevertheless, in the second column the number of non-equilibrium stages is lower than that predicted by the equilibrium model. By the other hand, when the mixture M4F1 is

Table 10
Design parameters for selected cases of conventional extractive distillation, M3F1.

	Q low		Q high	
	CDES, EQ	CDES, NEQ	CDES, EQ	CDES, NEQ
First column				
Number of stages	206	234	167	190
Feed stage	112	146	111	136
Extractant feed stage	57	82	55	79
Extractant flow rate (kmol/h)	562.9	691.9	582.4	775.2
Reflux ratio	2.7	2.6	2.7	2.6
Diameter (m)	2.5	2.8	2.5	2.6
Second column				
Number of stages	86	105	58	80
Feed stage	35	51	37	43
Reflux ratio	1.07	1.17	1.07	1.3
Diameter (m)	1.63	2.2	1.63	2.1
Total heat duty (MJ/h)	52,895.8	53,530.4	53,004.5	55,288.7

Table 11
Design parameters for selected cases of thermally coupled extractive distillation, M3F1.

	Q low		Q high	
	TCEDS, EQ	TCEDS, NEQ	TCEDS, EQ	TCEDS, NEQ
First column				
Number of stages	218	247	152	175
Feed stage	88	116	80	116
Extractant feed stage	46	70	43	65
Extractant flow rate (kmol/h)	704.3	778.4	720.7	778.4
Reflux ratio	2.23	2.55	2.3	2.55
Diameter (m)	2.35	2.6	2.34	2.6
Interlinking stages	120, 128	140, 141	110, 119	140, 140
Interlinking flow rate (kmol/h)	495.4	536.1	504.4	536.1
Second column				
Number of stages	46	29	46	30
Feed stage	46	29	46	30
Diameter (m)	1.56	1.8	1.58	1.8
Total heat duty (MJ/h)	48,827.6	52,887.4	49,309.1	52,972.9

separated, the number of non-equilibrium stages on the second column is more than twice the number of equilibrium stages. In this case, the component obtained as product in the second column is water, which has a relatively high composition in the feed stream.

6. Conclusions

A strategy for the optimal design of distillation sequences modeled with non-equilibrium stages has been proposed. The optimal design tool is based on a multiobjective stochastic approach coupled to a process simulator, which also considers the handling of constraints. Optimal designs of distillation sequences with both equilibrium and non-equilibrium models are compared in terms of energy requirements and number of stages, in order to find useful information that can lead to designs with best performance considering just the equilibrium model.

It has been found that for ordinary distillation in conventional sequences, the number of non-equilibrium stages required for a minimum in energy requirements is higher than the number of equilibrium stages, about 1.3–3.0 times, depending on the nature of the components of the mixture and the feed composition. In the case of conventional extractive sequences, optimal N_{NEQ}/N_{EQ} ratios are between 0.4 and 2.5. By the other hand, for ordinary and extractive distillation with thermally coupled sequences, similar patterns have been observed. Nevertheless, for some cases the number of non-equilibrium stages in the side rectifier is lower than the number of equilibrium stages.

Thus, the energy requirements obtained with the equilibrium model are reliable, since they are the same generated in the non-equilibrium model. However, the number of stages of equilibrium designs must be increased in order to have a better performance; as was mentioned previously. In summary, non-equilibrium designs can be obtained taking as a basis optimal equilibrium designs, correcting the number of stages by using the N_{NEQ}/N_{EQ} ratio. Such non-equilibrium designs may be taken as good approaches for a non-equilibrium optimal or as an initial point for a more rigorous optimization, reducing the number of iterations and the high computational effort required for optimizing with the non-equilibrium model.

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