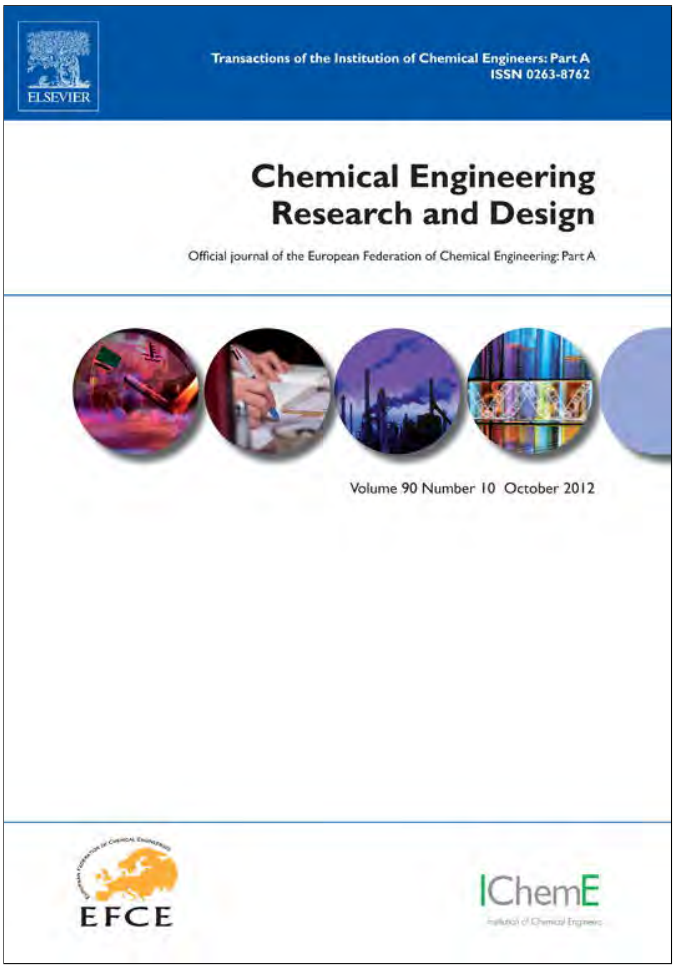

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Optimal design of distillation systems with less than $N-1$ columns for a class of four component mixtures

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A B S T R A C T

The optimal design of complex distillation systems is a highly non-linear and multivariable problem, with several local optimums and subject to different constraints. In addition, some attributes for the design of these separation schemes are often conflicting objectives, and the design problem should be represented from a multiple objective perspective. As a result, solving with traditional optimization methods is not reliable because they generally converge to local optimums, and often fail to capture the full Pareto optimal front. In this paper, a method for the multiobjective optimization of distillation systems, conventional and thermally coupled, with less than $N-1$ columns is presented. We use a multiobjective genetic algorithm with restrictions coupled to AspenONE Aspen Plus; so, the complete MESH equations and rigorous phase equilibrium calculations are used. Results show some tendencies in the design of intensified sequences, according to the nature of the mixture and feed compositions.

Keywords: Multicomponent distillation; Multiobjective optimization; Genetic algorithms; Pareto front

1. Introduction

Distillation is a widely used separation process, and, also, a very large consumer of energy. A large amount of research work has been done to improve the energy efficiency of distillation systems, in terms of either the design of optimal distillation schemes or improving internal column efficiency. Nowadays, the optimal design of multicomponent distillation systems remains one of the most challenging problems in process engineering (Kim and Wankat, 2004). The economic importance of distillation separations has been a driving force for the research in synthesis procedures for more than 30 years.

For the separation of an N-component mixture into N pure products, as the number of components increases the number of possible simple column configurations sharply grows up. Thereby, the selection of the best configuration for certain mixture was a very large and time consuming problem. This was one main reason that conducts researchers' work in searching general rules, heuristics, able to make a preliminary screening to select a lower number of alternatives to analyze. The selected configurations can be subsequently analyzed with a rigorous method to find the best sequence in terms of energy requirement (Nath and Motard, 1981; Nadgir and Liu, 1983).

At the moment, most of the studies have been focused on complete separation of N component mixtures using $N-1$ distillation columns, with a reboiler at the bottom and a condenser at the top of each column. Glinos and Malone (1989) show the application of thermodynamic principles to the synthesis of distillation sequences. While economic criteria are preferable for synthesis in the general case, the thermodynamic analysis has an advantage when heat integration of the distillation system is planned, or to further

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Nomenclature

Symbols

b	exergy function (Btu/h)
$[CO_2]_{Emiss}$	carbon dioxide emissions (kg/s)
DSC	conventional direct sequence
h	molar enthalpy (Btu/lb mol)
h_{proc}	enthalpy of steam delivered to the process (kJ/kg)
n	mole flow (lb mol/h)
NHV	the net heating value of a fuel (kJ/kg)
LW	the lost work in the system (kJ/kg)
λ_{Proc}	latent heat of steam delivered to the process (kJ/kg)
η	the thermodynamic efficiency
Q	reboiler heat duty (Btu/h)
Q_{Fuel}	amount of fuel burnt (kW)
S	molar entropy (Btu/lb mol R)
TAC	total annual cost ($/y)
T_0	temperature of the surroundings (R)
T_{FTB}	the flame temperature of the boiler flue gases (°C)
T_s	temperature of the system (R)
W_{min}	minimum work for the separation (Btu/h)
W_S	shaft work (Btu/h)
x	the number of atoms of carbon
y	the number of atoms of hydrogen

discriminates among configurations having comparable economic performance. Agrawal (2003) proposed an algorithm to systematically draw basic configurations by classifying them into different equivalence classes based primarily on whether column products were made as distillates, side draws or as bottoms. The number of configurations in each of these equivalence classes (i.e., the cardinality of that class) was evaluated using symmetry and simple combinatorial methods. The union of all these equivalence classes was called the set of "basic configurations". A basic configuration may therefore be defined as a configuration that uses $(N-1)$ columns to separate an N-component feed into N streams each of which is rich in one of the feed components, where each component is recovered in only one stream from only one location. The characteristic of this method is that it only generates basic configurations. The method was also extended to include corresponding thermally coupled configurations. On the other hand, an exhaustive combinatorial search (Rong et al., 2003; Fidkowski, 2006) is more complete. However, it is not compact, as it includes a very large number of configurations that use more than $(N-1)$ columns for an N-component feed when n is greater than three. These configurations, which we term "nonbasic" configurations, recover at least one component or mixture in at least two different locations, in such a way that sections cannot be grouped to make $(N-1)$ columns. The formulation of the search space has not enjoyed as much success in spite of earlier efforts. One reason for this was advanced by Westerberg (2004), that any formal mathematical or logical formulation of the search space can be developed only after drawing some configurations through a process of discovery, or by exploring the combinatorial of the system manually, and perhaps recognizing patterns underlying the space. Hence, if some structures are omitted (consciously or otherwise) in this phase of the work, the resulting formulation may or may not encompass the omitted structures. (Such incomplete enumeration of the search space will of course affect the final design whether or not a mathematical formulation is used). Therefore, in order to find optimal configurations it is essential that along with mathematical optimization methods, proper attention be paid to the search space formulation. Giridhar and Agrawal (2010a,b) focuses on formulating such a search space for multicomponent distillation, and describing some desirable properties of a good such search space. However, the use of less than $N-1$ columns for multicomponent distillation processes has not been extensively studied (Kim and Wankat, 2004; Méndez-Valencia et al., 2008). The problem of analyzing all possible sequences became more complex, when the use of the thermal coupling or side stream sequences was proposed (Errico et al., 2009a).

The optimization of a distillation column involves the selection of the configuration and operating conditions to minimize the total investment and operation cost (Yeomans and Grossmann, 2000). The optimization of a complex distillation system is usually characterized as a large problem size, since the significant number of strongly nonlinear equations results in serious difficulty to solve the model. At the moment, two principal approaches to optimize distillation columns sequence have been used: deterministic and stochastic.

Deterministic strategies tackled the problem considering the sequence as superstructures solved with MILP and MINLP approaches, considering reduced models (Aggarwal and Floudas, 1990; Andrecovich and Westerberg, 1985; Paules and Floudas, 1990; Viswanathan and Grossmann, 1993; Novak et al., 1996; Smith and Pantelides, 1995; Papalexandri and Pistikopoulos, 1996; Bauer and Stichlmair, 1998; Dunnebier and Pantelides, 1999; Caballero and Grossmann, 1999; Yeomans and Grossmann, 2000). Some of these methods can provide useful results; however, it would be desirable incorporate rigorous models in the synthesis procedures, in order to increase their industrial relevance and scope of application, particularly for nonideal mixtures. These rigorous MINLP synthesis models exhibit significant computational difficulties, such as the introduction of equations that can become singular, the solution of many redundant equations, and the requirement of good initialization points. The high levels of nonlinearities and nonconvexities in the MESH equations, thermodynamic equilibrium equations, and convergence difficulties are common problems. These difficulties translate into high computational times and the requirement of good initial guesses and bounds on the variables to achieve convergence. Also, these approaches just deal with one objective, total annual cost in most of the cases. Although Messac et al. (2003) present good alternatives for the solution of this problem with a multiobjective perspective using non-linear programming optimizers, clear limitations in the case of solving bi-objective problems are shown in Martínez et al. (2007).

In general, the optimal design of distillation systems is a highly non-linear and multiobjective problem, with the presence of both continuous and discontinuous design variables; also, the objective function used as optimization criterion is generally non-convex with several local optimums and subject to several constraints. Thereby, the problem must be tackled with a multiobjective strategy robust and reliable.

Multiobjective optimization techniques offer advantages over single-objective optimization techniques, because they can provide a solution with different trade-offs among different individual objectives of the problem; so, the decision

maker can select the best final solution for his/her particular needs. The principal advantage of using a multiobjective optimization technique is that just one run is required to obtain the Pareto front (Gutiérrez-Antonio and Briones-Ramírez, 2009).

Solving a multiobjective problem is, in general, associated with the construction of a Pareto front. Each point of this front represents one solution in the objective function space of the multiobjective problem. Therefore, given any pair of solutions as vectors of their objective function values, an improvement of one component involves a deterioration of the others. Thus, in the Pareto front there is no point that is better than another point (non-dominated points), and the rest of the objective space points are also dominated by one or more points (dominated points). The construction of a Pareto front can be very complex, depending on the nature of the multiobjective problem to be solved. The presence of multi-modal objective functions and non-convex constraints can complicate the task of the optimizer. Specifically, the use of optimizers based on non-linear programming can present problems to determine Pareto fronts, since non-linear programming depends on initial estimations, given its nature as a local optimizer for non-convex functions. These limitations are even greater for three-objective and multi-objective problems in general; therefore more research in that direction could be challenged.

On the other hand, stochastic optimizers deal with multimodal and non-convex problems in a very effective way to solve these limitations. Stochastic optimization algorithms are capable of solving, robustly and efficiently; the challenging multi-modal optimization problem, and they appear to be a suitable alternative for the design and optimization of complex separation schemes (Martínez-Iranzo et al., 2009). Among stochastic algorithms, genetic algorithms have shown their merits in large-scale parallelism search, approaching the global optimum quickly and steadily. Genetic algorithms are a well-established method, which has shown to have good performance (although it does not guarantee optimality).

A genetic algorithm is based on mimicking the principles of genetics, natural selection, and evolution; it allows a population composed of many individuals to evolve under specified selection rules to a state that maximizes the fitness, i.e., minimizes the cost function (Nguyen and Bagajewicz, 2010). Moreover, genetic algorithms have several features that make them attractive for solving optimization problems with modular simulators, where the model of each unit is only available in an implicit form (black-box model). First, due to the fact that they are based on a direct search method, it is not necessary to have explicit information on the mathematical model or its derivatives. Secondly, the search for the optimal solution is not limited to one point, but rather relies on several points simultaneously; therefore the knowledge of initial feasible points is not required, and such points do not influence the final solution (Teh and Rangaiah, 2003). Also, a major advantage of genetic algorithms (GA) over other stochastic techniques is the availability of several multiobjective techniques such as VEGA, MOGA, NSGA, Niche Pareto GA and NSGA-II (Vázquez-Castillo et al., 2009). Many studies have been carried out applying genetic algorithms to design in chemical engineering (Fraga and Matias, 1996; Leboreiro and Acevedo, 2004; Gómez-Castro and Segovia, 2008; Gutiérrez-Antonio and Briones-Ramírez, 2009; Miranda-Galindo et al., 2011).

In this work, we implemented a multiobjective genetic algorithm with constraints to obtain the Pareto front of complex distillation sequences. We take an alternative space of 31 arrangements that use less than $N-1$ columns, for quaternary distillations of those reported by Errico et al. (2009a,b). The optimal designs of these arrangements are obtained using a multiobjective genetic algorithm with restrictions (Gutiérrez-Antonio and Briones-Ramírez, 2009), which is coupled to the AspenONE Aspen Plus for the rigorous evaluation of the objective and constraint functions. To the best of our knowledge, multiobjective stochastic methods have not been reported for optimal design of a complete subspace of alternatives with less than $N-1$ columns for quaternary distillations. Numerical performance of this strategy has been tested in the design of columns with several mixtures, in order to examine the effect of the relative volatilities of feed mixtures. Results allow presenting trends for selecting the best configuration among those studied according to the nature of the mixture and feed composition.

2. Pareto front and optimization strategy

Decision making in the Chemical Process Industry involves selecting from a large list of potential solutions. The selection is virtually always based on multiple independent and often conflicting criteria, including cost, risk, safety, reliability and environmental impact. As a result, there is seldom a single, numerically optimal, answer. To make an informed decision, the decision-maker needs to identify the inherent tradeoffs among the various criteria. These trade-offs can be represented through a Pareto set of options (Deb et al., 2002), wherein no member of the set is strictly better than any other member of the set. The generation of Pareto sets through formal, rigorous multi-objective optimization involves the solution of a combinatorially explosive number of subproblems, each one of which may be non-convex and hard to solve (Siirola et al., 2004).

Classically, optimization focuses primarily on problems that can be evaluated and ranked based on a single metric. Solving such a single-objective optimization problem involves identifying the one solution (vector of values for the problem decision variables) that maps to the best value of the objective possible, while simultaneously satisfying all constraints. There exists a plethora of algorithms and approaches (Edgar et al., 2001) that are effective in solving single objective problems, including gradient based algorithms (generalized reduced gradient, sequential quadratic programming), stochastic algorithms (simulated annealing, tabu search), and population-based algorithms (genetic algorithms). However, many of the problems that are of key interest to the design decision maker are evaluated using multiple conflicting goals (Papalambros and Wilde, 2000). The solution to this class of multiple objective problems cannot be represented as a single point; rather, it is the set of feasible points that represent the best trade-offs among the various objectives. By definition, all members of this set are mathematically optimal: none of the set members is better than any other member in all objectives. It is thus up to the business decision maker to apply information external to the problem, and/or based on the unanticipated shape of the tradeoff surface to select the final, single solution to implement. More formally, multi-objective optimization entails the discovery of the Pareto-optimal set of non-dominated solutions.

For problems where the goal is to minimize all objectives, the dominance Pareto relationship (Grosan et al., 2006) is defined as:

Definition 1. (Pareto dominance)
Consider a maximization problem. Let z, w be two decision vectors (solutions) from the definition domain. Solution z dominates w (also written as $z \succ w$) if and only if the following conditions are fulfilled:

$$f_i(z) \geq f_i(w), \quad \forall i = 1, 2, \ldots, nobj \tag{1}$$

$$\exists j \in \{1, 2, \ldots, nobj\} : f_j(z) \rangle f_j(w). \tag{2}$$

where $nobj$ denotes the number of objectives. That is, a feasible vector z is Pareto optimal if no feasible vector w can increase some criterion without causing a simultaneous decrease in at least one other criterion. The Pareto optimal set is the collection of all points for which there exists no other feasible point that dominates it.

For the specific problem of optimization of the arrangements with less than $N-1$ columns for quaternary distillations, our goal is generated the Pareto front of optimal designs; this set of optimal designs is integrated for all distillation sequences from minimum reflux ratio to minimum number of stages, and all designs between these extremes. The analysis of the designs that integrated the Pareto front can bring useful information about some tendencies of these intensified sequences. Thereby, obtaining the set of optimal designs of the intensified sequences involves the minimization of heat duty, Q_i, and the number of stages in each column of the sequence, N_i, can be formulated as:

$$\begin{aligned} &Min(Q_i, N_i) = f(Q_i, R_i, N_i, N_{j,i}, N_{s,i}, N_{F,i}, F_{j,i}) \\ &\text{subject to} \\ &\vec{y}_{k,PS} \geq \vec{x}_{k,PS} \end{aligned} \tag{3}$$

where R_i is the reflux ratio in the column i, N_i is the total number of stages of column i, $N_{j,i}$ is the stage of the flow of interconnection stream j in the column i, $N_{s,i}$ is side stream stage in column i, $N_{F,i}$ is the feed stage in column i, $F_{j,i}$ is the flow of interconnection stream j in column i, $\vec{x}_{k,PS}$ and $\vec{y}_{k,PS}$ are the vectors of required and obtained purities of component k in their product stream PS. It is worth to mention that the formulation of this minimization problem is general, and it can apply for each sequence studied in this work, just considering the variables that are presented in the particular sequence.

Thereby, the variables in competition, Q_i and N_i, are involved as the objectives in the optimization problem. Also, this minimization takes place considering the fulfilment of the constraints of the problem, these constraints limit the minimization problem: we are looking for all the sequences that integrate the Pareto front, which minimizes as the heat duty as the number of stages in each column of the sequence but satisfying the purities required in each product stream.

The stochastic strategy implemented was presented previously (Gutiérrez-Antonio and Briones-Ramírez, 2009), and it is briefly explained next. For further details the reader is referred to the work. The optimization strategy considers the simultaneous optimization of several objectives subjects to several constraints; both constraints and objectives can be as integer as continuous. The strategy conducts the optimization process, using Aspen Plus for the evaluation of the individuals, distillation sequences, generated as is explained next. The stochastic strategy needs one individual physically feasible to begin the optimization process; the meaning of physically feasible implies that, for a distillation column, the feed stage must be minor than the total number of stages, and the reflux ratio is greater than zero. With this initial individual, the genetic algorithm generates n_{ind} individuals varying all the variables of design of the distillation scheme (as feed location, interconnection flow streams, interconnection stage location, among others); in this way, the algorithm has n_{ind} designs of the distillation schemes. These schemes are sent to Aspen Plus, one by one, in order to simulate the distillation scheme and get the values of the objectives and constraints of each individual. With the information of the objectives and constraints, the genetic algorithm classifies the individuals according with the number of constraints satisfying, and inside each subgroup according with the fitness value. Then, the best individuals are chosen to be the parents of the new generation, and so; until the total number of generations has been reached. Then integration between the rigorous simulation and the stochastic optimization strategy is made with technology ActiveX.

For this minimization, the implemented multiobjective algorithm is based on the NSGA-II (Deb, 2001; Deb et al., 2002), and the constraints are handling using a slight modification of the work of Coello-Coello (2000). This modification is as follows: the entire population is divided into sub-populations using, as criterion, the total number of satisfied constraints, n_{const}. Thus, the best individuals of the generation are those that satisfied all n_{const} constraints, and they are followed by the individuals that only satisfying $n_{const}-1$, and so on. Within each sub-population, individuals are ranked using the NSGA-II, but considering now as another objective function to minimize the difference between the required and obtained constraints. Next, dominance calculation of each subgroup is carried out as follows:

$$\text{dominance}\{Q_i, N_i, \text{minimum}[0, (\vec{x}_{k,PS} - \vec{y}_{k,PS})]\} \tag{4}$$

According to Eq. (4), in each subgroup we are minimizing as the original objective functions as the difference between the required and obtained constraints. In this way, the minimization takes into account as the objectives as the constraints of the problem.

3. Cases of study

As was mentioned before, we selected as study a subset of 31 configurations that use less than $N-1$ columns, for quaternary distillation, proposed by Errico et al. (2009a,b). These configurations were divided in three principal groups:

1. Distillation sequences with less than $N-1$ columns without thermal coupling.
2. Distillation sequences with less than $N-1$ columns with thermal coupling.
3. Single distillation columns with two side streams.

Also, each group is divided in subgroups (SG). The group 1 is divided in the following subgroups depending on the product stream that is fed to the second column:

- SG1: distillate stream of the first column is fed to the second one.
- SG2: side stream of the first column is fed to the second one.
- SG3: bottom stream in the first column is fed to the second one.

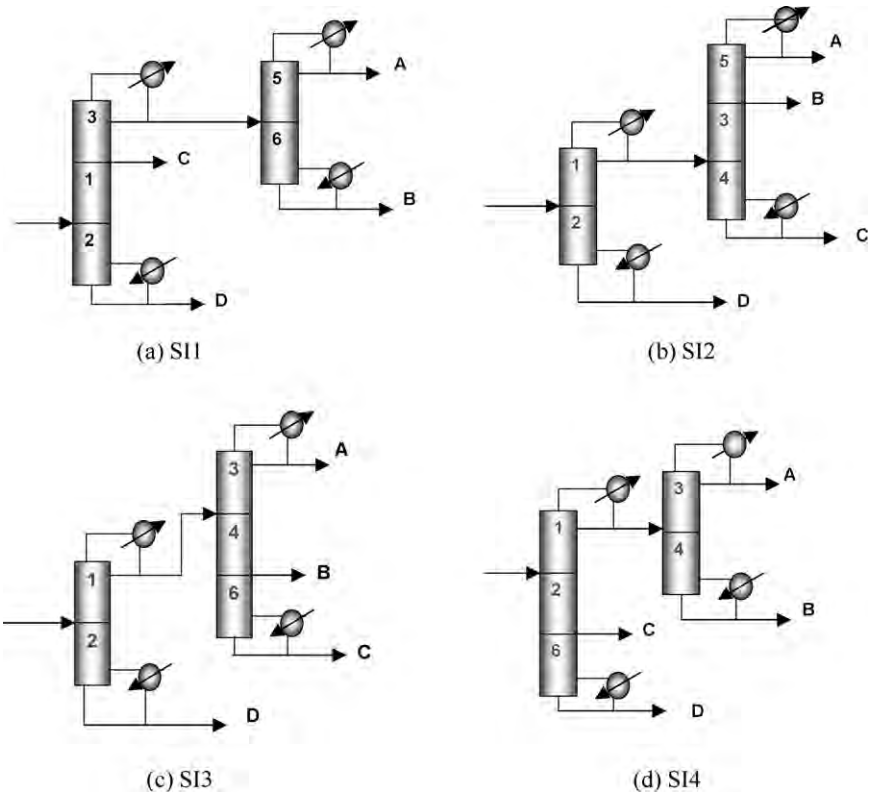

Fig. 1 – Intensified sequences of subgroup 1, SG1.

On the other hand, in the group 2 there are 5 subgroups according to the location of the interconnection streams and the type of second column:

- SG4: interconnection streams at the top of the first column linked to a single column.
- SG5: interconnection streams at the middle of the first column linked to a side rectifier.
- SG6: interconnection streams at the middle of the first column linked to a side stripper.
- SG7: interconnection streams at the middle of the first column linked to a single column.
- SG8: interconnection streams at the bottom of the first column linked to a single column.

Finally, the group 3 contains the unique distillation column with two side streams, subgroup SG9. Figs. 1–9 show the 9 subgroups of the intensified distillation sequences.

To compare the performance of the sequences with less than $N-1$ columns, four quaternary mixtures with different volatility values and feed compositions were considered, Table 1. These case studies, in composition and volatilities, are similar to those previously analyzed by Kim and Wankat (2004) and Méndez-Valencia et al. (2008) for systems with less than $N-1$ columns. The required purities were 98.7%, 98%, 98% and 98.6% for components A, B, C and D, respectively. For all mixtures, the feed flowrate is 45.36 kmol/h and the feed is introduced in the column as saturated liquid. The design pressure for each separation was chosen to ensure the use of cooling water in the condensers; and they were 4.82, 4.77, 6.59 and 2.34 atm, for mixtures M1, M2, M3 and M4, respectively.

It is well known that the composition in the feed stream affects the design parameters and energy consumption; for that reason, different feed compositions and volatilities values were considered. Phase equilibrium for liquid of these mixtures is calculated with the Chao-Seader model for mixtures M2 and M3, mixtures of alcohols and isomers. The Chao-Seader correlation is usually recommended for hydrocarbon mixtures operating at low or medium pressure (Aspen Plus Manual, 2007; Errico et al., 2009b), and UNIQUAC model for mixtures M1 and M4. As was mentioned before, the simulations of all sequences are performed in Aspen Plus in

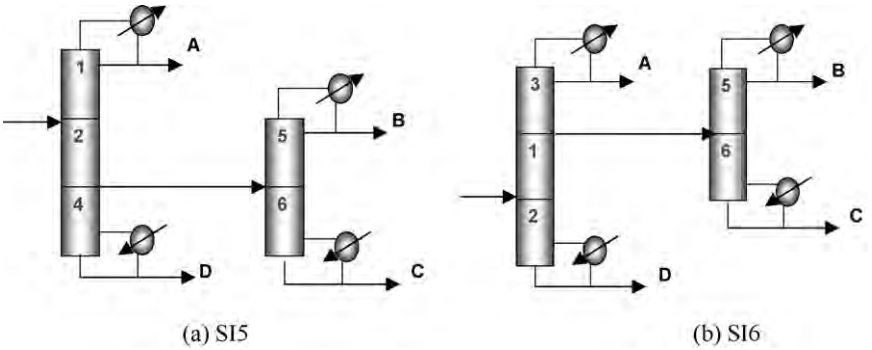

Fig. 2 – Intensified sequences of subgroup 2, SG2.

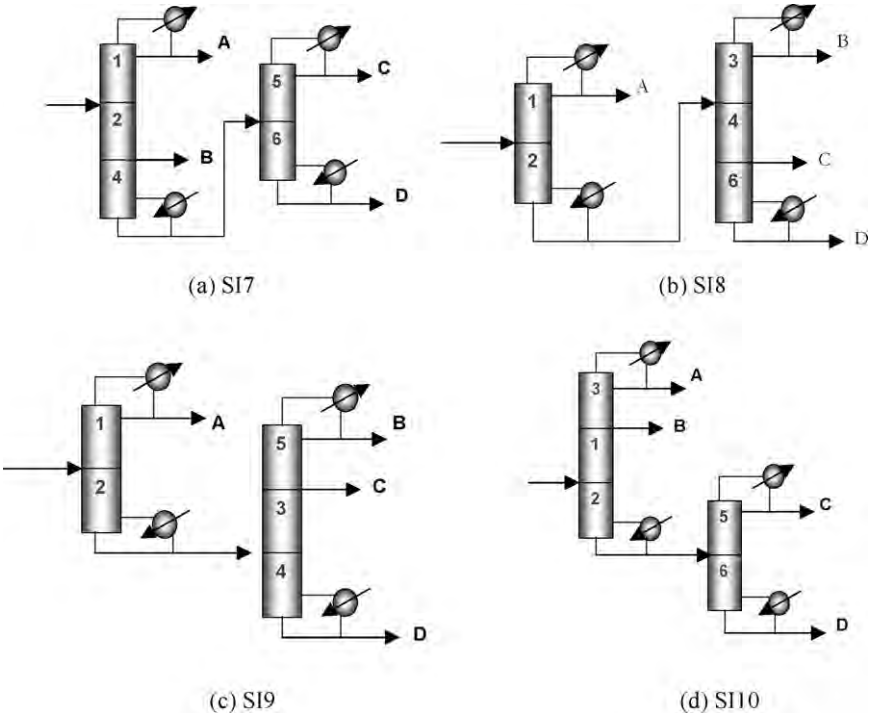

Fig. 3 – Intensified sequences of subgroup 3, SG3.

order to get the values of objectives and constraints; thereby, the phase equilibrium calculations are performed in Aspen Plus. The phase vapor is model with Rendlich Kwong Soave model.

For the complex distillation sequences we used 2500 individuals and 40 generations as parameters of the genetic algorithm, with 0.80 and 0.05 of crossover and mutation fraction. These parameters were obtained through a tuning process, where several runs of the algorithm were performed with different numbers of individuals and generations. It worth mentioning that during the simulation process, an infinite heat duty is assigned by the algorithm to the individual which simulation converges with errors; if the simulation does not converge; the algorithm also settles as zero the values of purities and recoveries. Approximately, the time required to perform the optimization of each sequence is about 6 h; once the tuning process has been completed. For more detailed information about this algorithm and its link to Aspen Plus, the reader is referred to the original work (Gutiérrez-Antonio and Briones-Ramírez, 2009).

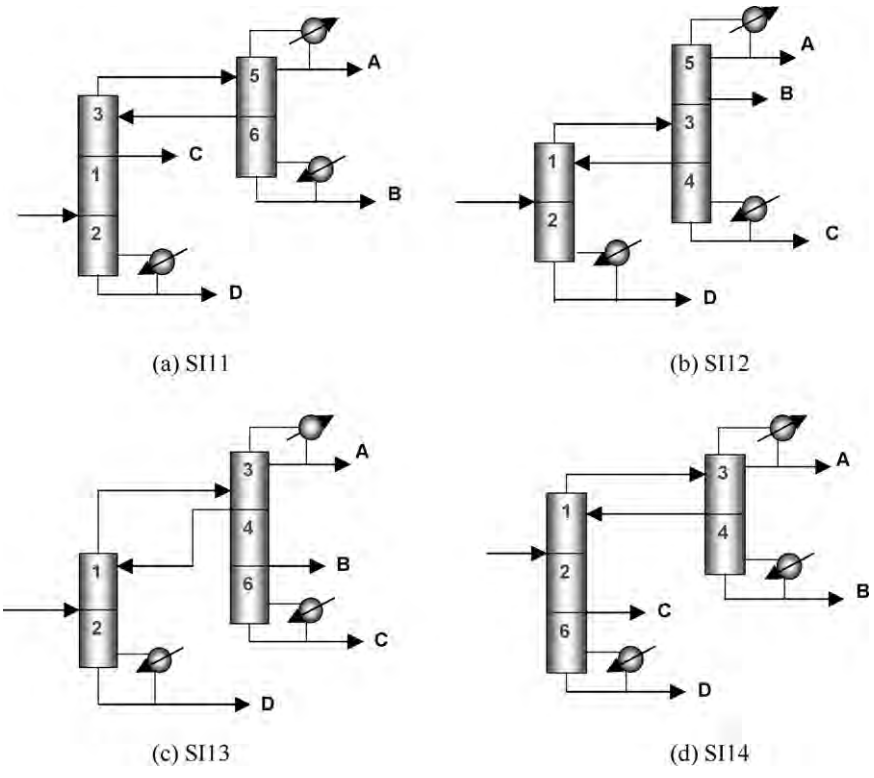

Fig. 4 – Intensified sequences of subgroup 4, SG4.

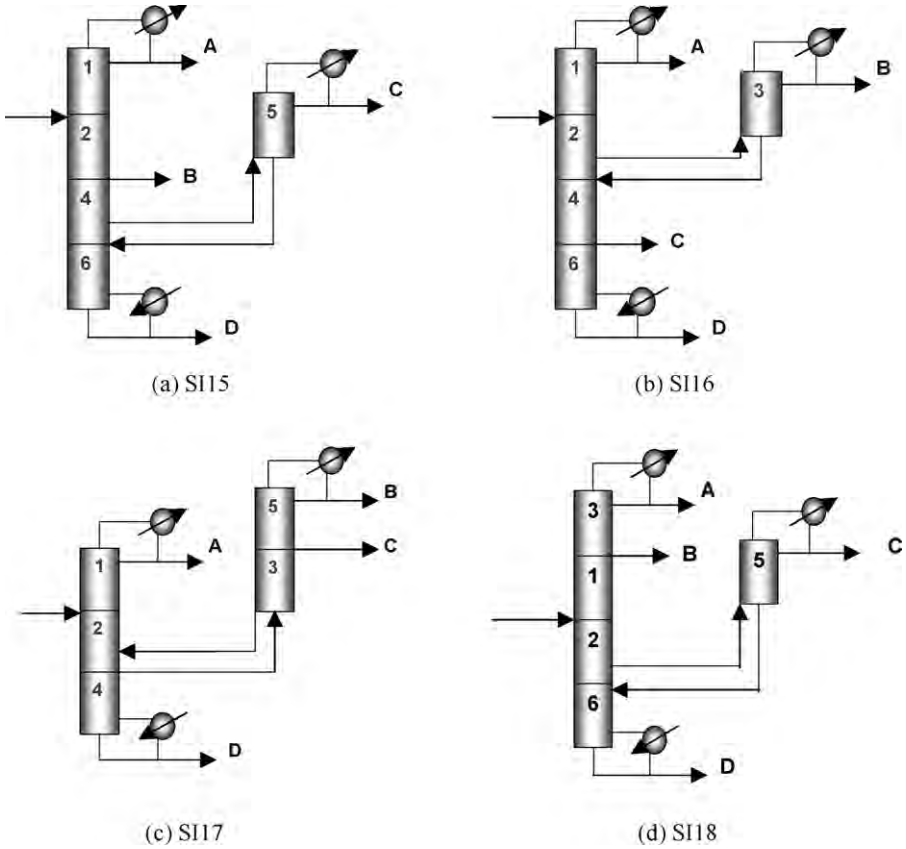

Fig. 5 – Intensified sequences of subgroup 5, SG5.

4. Results

In this section we present the optimal designs that integrate the Pareto fronts for all mixtures and studied sequences. The resulting Pareto fronts are integrated by the optimal designs that represent the best trade-offs between the considered objectives: number of stages and the heat duty in each column. In order to visualize better the results, we decide to group the

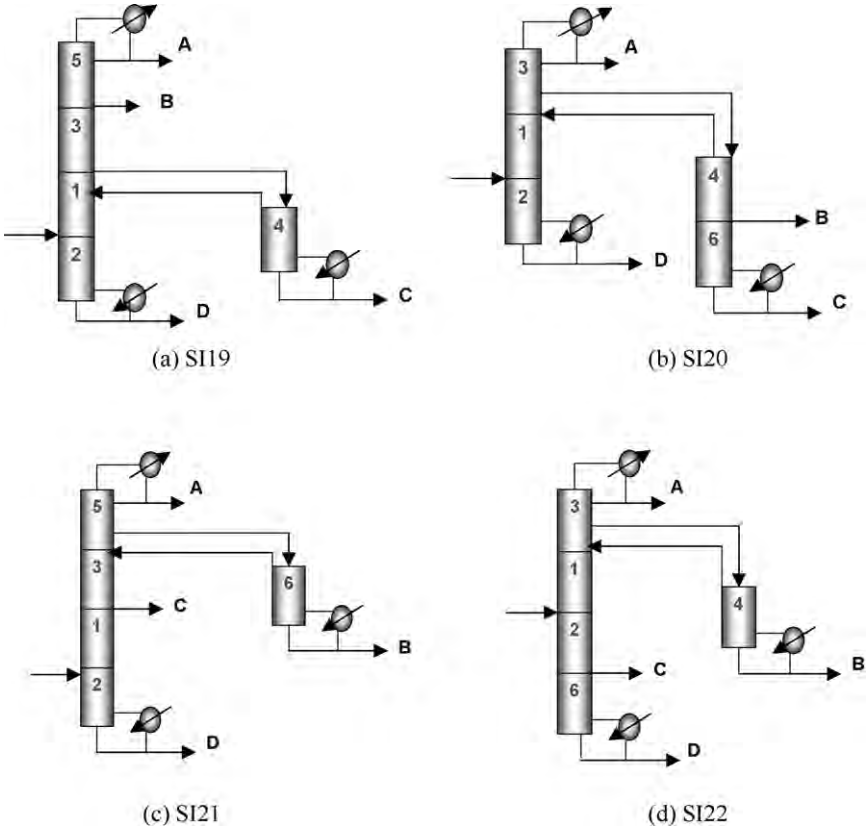

Fig. 6 – Intensified sequences of subgroup 6, SG6.

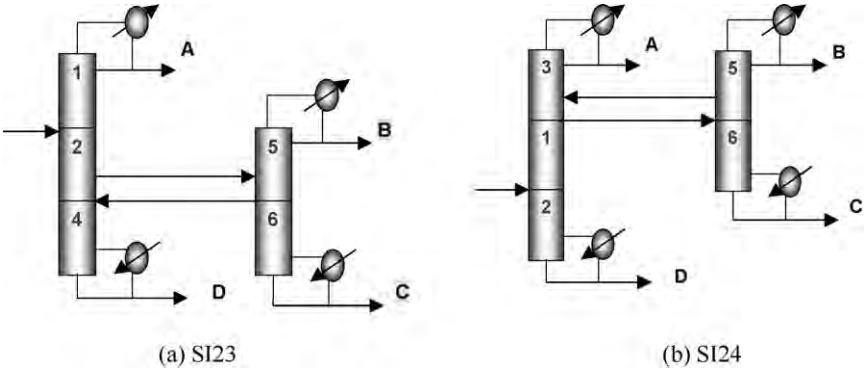

Fig. 7 – Intensified sequences of subgroup 7, SG7.

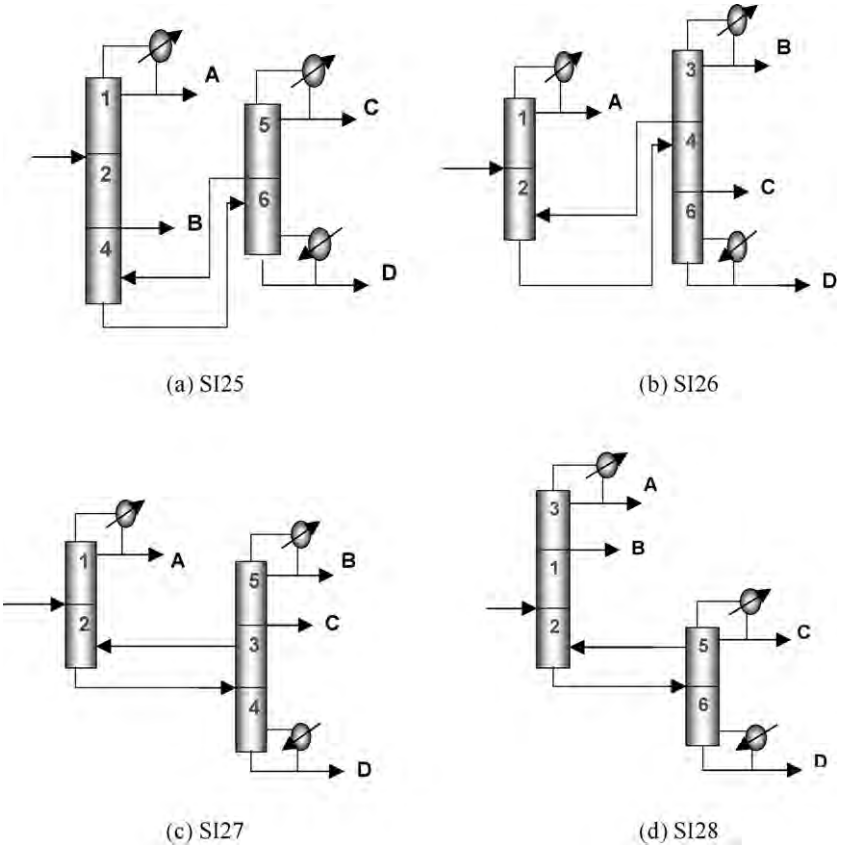

Fig. 8 – Intensified sequences of subgroup 8, SG8.

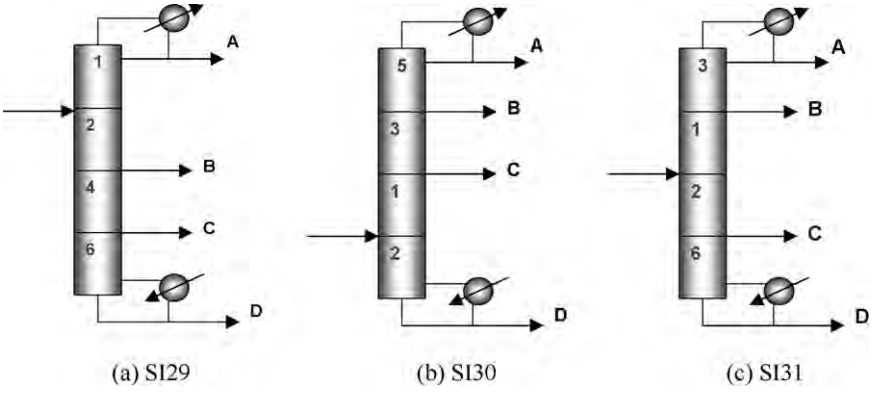

Fig. 9 – Intensified sequences of subgroup 9, SG9.

Table 1 – Selected quaternary mixtures.

Mixture	Components	x_i	$\alpha_{i,H}$	$\alpha_{i,j}$	T (°C)	ΔT (°C)
M1	A: isobutane	0.40	6.07		−11.72	
	B: n-butane	0.10	4.68	1.29	−1.00	10.72
	C: n-pentane	0.10	1.91	2.45	36.07	37.07
	D: iso-hexane	0.40	1.00	1.91	60.01	23.94
M2	A: n-butane	0.30	27.67		−1.00	
	B: n-pentane	0.40	11.44	2.42	36.07	37.07
	C: n-hexane	0.25	4.94	2.32	68.73	32.66
	D: n-octane	0.05	1.00	4.94	125.68	56.95
M3	A: n-butane	0.05	10.07		−1.00	
	B: n-pentane	0.45	4.5	2.24	36.07	37.07
	C: n-hexane	0.45	2.08	2.16	68.73	32.66
	D: n-heptane	0.05	1.00	2.08	98.43	29.70
M4	A: isobutanol	0.05	5.77		107.66	
	B: 1-pentanol	0.25	3.05	1.89	137.80	30.14
	C: 1-hexanol	0.40	1.78	1.72	157.40	19.60
	D: isoheptanol	0.30	1.00	1.78	172.01	14.61

heat duty of the sequence, instead of plotting this variable for each column. Thereby, the Pareto fronts show the number of stages in each column and the total heat duty of the sequence. Also, with the information generated by the Pareto front, we calculate some other variables as thermodynamic efficiency, total annual cost and CO_2 emissions in order to evaluate them integrally.

4.1. *Mixture 1*

The first mixture in our analysis is a mixture of hydrocarbons; the intermediate components, B and C, are of normal chain, and the light, A, and heavy, D, components are isomers. The feed composition contains a low concentration of the two intermediate components (20%). The relative volatilities of the adjacent components indicate that the harder separation is performed between isobutane, A, and n-butane, B; the easiest cut includes the two intermediate components: n-butane, B, and n-hexane, C.

Figs. 10–12 show the Pareto front of distillation sequences with less than $N-1$ columns: without thermal coupling, with thermal coupling, and the single distillation column, with two side streams, respectively. Now, from Fig. 10 we observe that the lower energy consumptions are shown by sequence SI8,

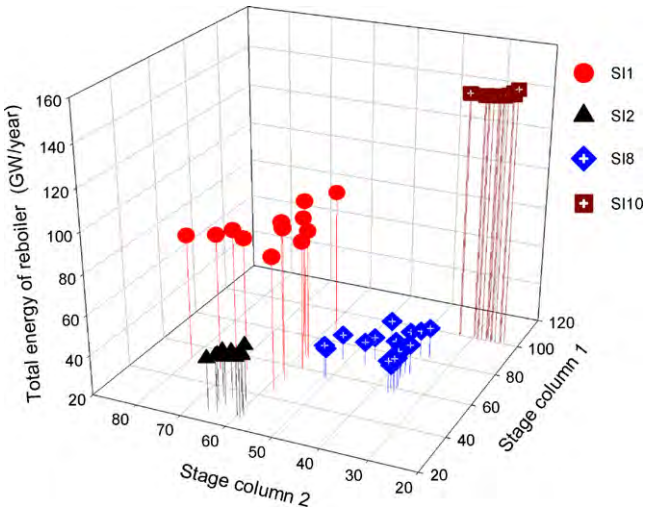

Fig. 10 – Pareto front of sequences with less than N – 1 columns without thermal coupling, mixture M1.

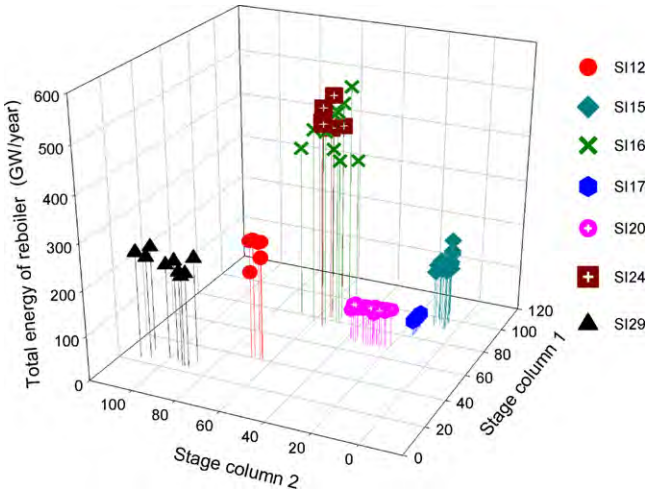

Fig. 11 – Pareto front of sequences with less than N – 1 columns with thermal coupling, mixture M1.

without thermal coupling, where one of the more abundant components is separated in the first column, as a top product. The sequence that follows this one is the SI2, without thermal coupling, which also separates one of the more abundant components in the first column, as a bottom product. The energy consumption of the sequence SI8 is very similar to the heat duty of the sequence SI17, with thermal coupling, as shown

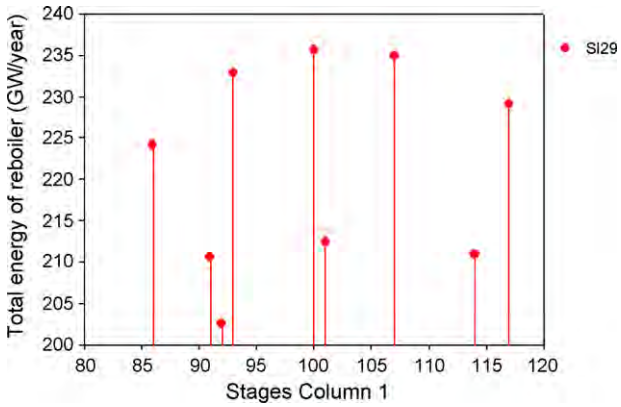

Fig. 12 – Pareto front of sequences with less than N – 1 columns with single distillation column, mixture M1.

in Fig. 11. In sequence SI17, the two abundant components are separated in the first column. Sequences SI2, SI8 and SI17 have in common the fact that one of the more abundant components is first separated in the first column from the rest of the mixture. Moreover, the minimum energy consumption is observed when the hard cut is performed in the first column; in other words, the hard cut is performed better in a sequence that separates one of the components of the hard cut from the rest of the mixture, instead of from the adjacent component. That is the reason why the sequences SI5 in SG5, and SI23 in SG7 have the worst performance in terms of energy consumption (in that sequences one component is separated from the adjacent par instead of being separated or the rest of the mixture). It is worth of mention that in spite of the similarity between sequences SI17 and SI23, the design of this last one generates a remixed of the intermediate components, increasing the heat duty. In particular, the sequence SI29, two side streams column, requires more than six times the energy consumed by the sequence SI8, even when the number of stages is quite similar, as shown in Fig. 12.

From these figures we observe that the minimum energy consumption and greater thermodynamic efficiency are presented in one sequence of two columns without thermal coupling, SI8; however, the thermally coupled ones are not so far from this minimum value (SI17). This result is quite unexpected, due to the well known behavior of columns with thermal coupling in terms of energy savings (Fonyó et al., 2001); however, it is important to remark that this observation applies to sequences with $N-1$ columns, and not for less than $N-1$. Nevertheless, both sequences have similar energy consumptions but sequence SI17 has a minor number of stages and a reflux ratio 10 times bigger than that presented in sequence SI8.

Table 2

shows thermodynamic efficiency, total annual cost and CO_2 emissions and the design variables for the sequence with minimum energy consumption of each group and the Direct Conventional Sequence (DCS). From this table we can observe that thermodynamic efficiency is greater in sequence SI8, followed by trains SI17 and SI2, the same tendency was found in the heat duties. As consequence, in this sequence SI8 the CO_2 emissions and total annual costs are lower.

For this particular case study, we see that if one of the components of the hard cut is in major proportion, choose the sequence that allows separating one of the components, the most abundant, of the harder cut from the rest of the mixture; in this way, the hardest cut is performing more efficiently. In the sequences of this type, the minimum energy consumption can be found in those without thermal coupling, considering similar number of stages.

4.2. *Mixture 2*

The second mixture in our analysis is another mixture of hydrocarbons; the light, A, intermediates, B and C, and the heavy component D are of normal chain. The feed composition contains a concentration of the light component of 30% (the second more abundant) and the heavy component of 5% (the less abundant). The relative volatilities in Table 1 indicate that the separations are in general easy. In this case, the easiest cut is among n-hexane (C)/n-octane (D); the less easy includes n-pentane (B), and n-hexane (C).

Fig. 13 shows the Pareto front of distillation sequences with less than $N-1$ columns without thermal coupling, with

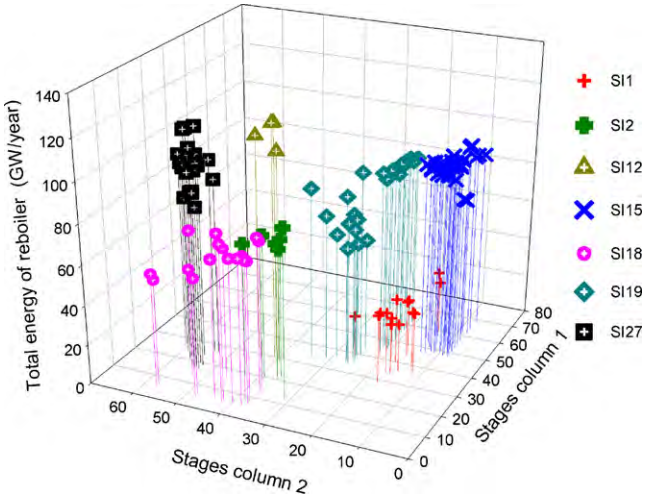

Fig. 13 – Pareto front of sequences with less than $N-1$ columns, mixture M2.

thermal coupling, and also with the single distillation column. Now, from Fig. 13 we observe that the lower energy consumptions are showed by sequence SI1 (only if it is compared with others intensified sequences, because for this mixture the minimum energy consumption is obtained in SC1), without thermal coupling, where the separation cut of the mixture, between column 1 and 2, is B/C. In the group of thermally coupled schemes, the sequence that follows in energy consumption is SI18 and SI19, where, again, the separation cut of the mixture, between column 1 and 2, is B/C. In these schemes, the common fact is that the separation of the hard cut is performed in the first column. As was observed in mixture M1, for mixture M2 the lower energy consumption is observed in a sequence without thermal coupling. In this mixture M2, thermally coupled sequences are not so close to the minimum energy consumption, SI1; in fact, the energy requirements are at least twice of those reported in the sequences without thermal coupling. Also, the single distillation column, SI31, has the higher heat duty of all sequences for mixture M2.

It seems that when the configuration of the sequence allows having the cut B/C to pass from first column to the second one the performance of separation is better. According to the results, the best option is considering the lower proportion component along with its adjacent component as a pseudo component, and performing the separation of this pseudo component from the rest of the mixture. Then, the pseudo component is purified in the first sequence. That is the reason why the sequences SI5, SG2, SI10, SG3, SI12, SG4, SI27, SG8, and SI15, SG5, have the worst performance in terms of energy consumption. Nevertheless, sequence SI31, SG9, requires more than ten times the energy consumed by sequence SI1, even when the number of stages of sequence SI31 is greater than the stages of scheme SI1.

From Fig. 13 we observe that the minimal energy consumption is presented in one sequence of two columns without thermal coupling, SI1; however, the thermally coupled ones are not so far from this minimum value, for instance SI8.

Table 3 shows thermodynamic efficiencies, total annual cost and CO_2 emissions for the sequences of all groups. From this table we can observe that thermodynamic efficiency is greater in sequence SC1, followed by trains SI1, as a consequence, in this sequence SC1 the CO_2 emissions and total annual costs are lower.

Table 2 – Summary of Pareto front for the intensified sequences, M1.

Intensified sequences	Group	Number sequence	Number column	Total stages	Feed stage	Side stage 1	Side stage 2	Vapor stage	Liquid stage	Reflux ratio	Diameter (m)	Total energy of reboiler (GW/year)
		DCS (conventional Direct sequence)	1	100	47	–	–	–	–	4.52	0.88	9.23
			2	25	11					5.75	0.54	2.66
			3	30	12					8.00	0.75	3.89
		SI1	1	44	29	16	–	–	–	37.33	3.07	76.03
	SG1		2	58	34	–				5.85	0.97	10.66
		SI2	1	35	15	–	–	–	–	2.84	1.04	10.47
2 columns			2	66	49	49				19.97	1.78	32.91
	SG2	SI5	1	75	19	46	–	–	–	72.12	2.18	41.63
			2	46	15	–				59.49	0.85	8.07
		SI8	1	59	30	–	–	–	–	6.03	0.98	11.62
	SG3		2	37	14	14				51.28	1.48	20.72
		SI10	1	101	40	80	–	–	–	85.43	4.30	136.12
			2	30	14	–				11.20	0.75	5.47
	SG4	SI12	1	40	17	–	–	–	–	166.73	0.87	7.37
			2	69	–	50		58	63	–	5.21	187.39
		SI15	1	104	14	60	–	75	13	78.41	4.23	126.10
			2	8	–	–		–	–	–	0.26	–
		SI16	1	104	61	57	–	42	–	204.98	7.99	321.64
	SG5		2	53	–	–		–	32	–	0.21	–
		SI17	1	81	29	63	–	95	–	5.41	1.93	33.89
Coupled			2	15	–	–		–	62	–	1.46	–
		SI18	1	29	–	–	–	–	–	–	0.40	–
			2	80	54	76		66	49	101.51	4.94	160.36
	SG6	SI20	1	69	58	46	–	31	–	42.70	2.76	63.82
			2	38	–	–		–	31	–	0.65	–
	SG7	SI24	1	85	42	35	–	40	124	5.63	5.63	194.99
			2	69	–	58		63	3.6	0.41	0.41	–
1 column	SG9	SI29	1	92	24	50	65	–	–	127.8	5.78	202.54

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Table 2 (*Continued*)

Intensified sequences	Group	Number sequence	Number column	Molar flow of A	Molar flow of B	Molar flow of C	Molar flow of D	Molar flow of liquid	Molar flow of vapor	Thermodynamic efficiency	CO_2 emissions	Total annual cost
						(Mton/year)				η	(Mton/year)	(MDD/year)
2 columns		DCS (conventional Direct sequence)	1 2 3	25.18	7.13	8.25	39.41	–	–	13.17	0.35	0.99
	SG1	SI1	1 2	19.73	4.70	5.75	29.30	–	–	3.17	0.89	3.16
		SI2	1 2	19.84	4.64	5.70	29.29	–	–	6.79	0.41	1.72
	SG2	SI5	1 2	19.84	4.61	6.71	28.13	–	–	5.4	0.58	2.06
	SG3	SI8	1 2	19.82	4.63	5.76	29.28	–	–	8.62	0.33	1.33
		SI10	1 2	19.79	4.74	5.80	29.07	–	–	1.91	1.47	5.06
Coupled	SG4	SI12	1 2	25.14	7.08	8.25	34.77	53.68	–	1.57	1.81	6.39
	SG5	SI15	1 2	25.29	7.10	8.19	34.59	–	12.37	2.05	1.37	4.57
		SI16	1 2	25.12	7.06	8.30	34.77	–	15.07	0.81	3.49	10.85
		SI17	1 2	25.20	7.02	8.18	34.82	–	301.25	7.68	0.37	1.46
		SI18	1 2	19.70	5.80	6.95	29.25	–	32.30	0.79	1.74	5.57
	SG6	SI20	1 2	25.19	7.12	8.12	34.78	–	77.95	3.83	0.65	2.57
	SG7	SI24	1 2	25.08	7.20	8.25	39.46	12.44	–	1.32	2.14	6.85
1 column	SG9	SI29	1	25.29	6.02	5.98	25.13	–	–	1.29	2.2	6.88

Table 3 – Summary of Pareto front for the intensified sequences, M2.

Intensified sequences	Group	Number sequence	Number column	Total stages	Feed stage	Side stage 1	Side stage 2	Vapor stage	Liquid stage	Reflux ratio	Diameter (m)	Total energy of reboiler (GW/year)
2 columns		DCS (conventional Direct sequence)	1	27	13	–	–	–	–	2.23	0.69	4.76
			2	21	10					1.56	0.80	4.55
			3	14	7					0.44	0.54	2.06
	SG1	SI1	1	39	10	21	–	–	–	3.69	1.34	17.01
			2	26	10	–				1.89	0.60	3.94
		SI2	1	28	15	–	–	–	–	1.05	1.06	11.02
			2	39	25	25				35.87	2.30	47.70
	SG2	SI5	1	42	14	15	–	–	–	64.15	2.51	52.00
			2	20	15	–				10.94	1.64	26.53
	SG3	SI8	1	13	7	–	–	–	–	6.55	0.98	10.32
			2	35	18	18				8.87	1.46	19.79
		SI10	1	33	23	14	–	–	–	44.26	2.56	58.94
			2	15	7	–				0.52	0.46	2.14
Coupled	SG4	SI12	1	30	9	–	–	–	–	77.68	0.88	7.62
			2	40	–	19		10	7	–	3.48	97.11
	SG5	SI15	1	57	10	17	–	28	21	51.42	3.15	73.80
			2	8	–	–		–	–	–	0.54	–
		SI18	1	6	–	–	–	–	–	–	0.50	–
			2	57	23	22		34	16	38.07	2.57	54.16
	SG6	SI19	1	37	30	18	–	24	–	52.60	2.59	54.98
			2	27	–	–		–	17	–	0.45	–
	SG8	SI27	1	22	11	–	–	–	–	35.55	0.73	–
			2	55	–	49		51	30	–	3.19	77.63
1 column	SG9	SI31	1	101	24	50	65	–	–	134.1	5.972	212.36

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Table 3 (*Continued*)

Intensified sequences	Group	Number sequence	Number column	Molar flow of A	Molar flow of B	Molar flow of C	Molar flow of D	Molar flow of liquid	Molar flow of vapor	Thermodynamic efficiency	CO_2 emissions	Total annual cost
						(Mton/year)				η	(Mton/year)	(MDD/year)
2 columns		DCS (conventional Direct sequence)	1 2 3	18.73	29.64	21.44	4.88	–	–	16.19	0.4	0.57
	SG1	SI1	1 2	14.68	24.48	18.20	4.37	–	–	10.46	0.53	0.94
		SI2	1 2	14.43	24.33	18.39	4.85	–	–	4.20	1.30	2.16
	SG2	SI5	1 2	14.62	60.64	18.25	4.49	–	–	1.63	2.19	2.99
	SG3	SI8	1 2	14.58	24.46	18.32	4.43	–	–	7.59	0.73	1.18
		SI10	1 2	14.49	24.31	18.46	4.55	–	–	4.09	1.35	2.15
	SG4	SI12	1 2	18.96	29.66	21.05	4.35	25.26	–	2.47	2.31	3.65
	SG5	SI15	1 2	18.97	29.62	21.23	4.19	–	33.82	3.09	2.10	2.94
Coupled		SI18	1 2	14.45	24.29	18.30	3.66	–	40.89	3.73	1.47	2.16
	SG6	SI19	1 2	18.46	29.76	21.74	4.22	–	35.41	3.85	1.62	2.25
	SG8	SI27	1 2	14.75	24.20	17.96	3.67	–	49.82	2.59	2.11	3.08
1 column	SG9	SI31	1	25.29	6.03	5.96	25.14	–	–	3.5	1.73	2.43

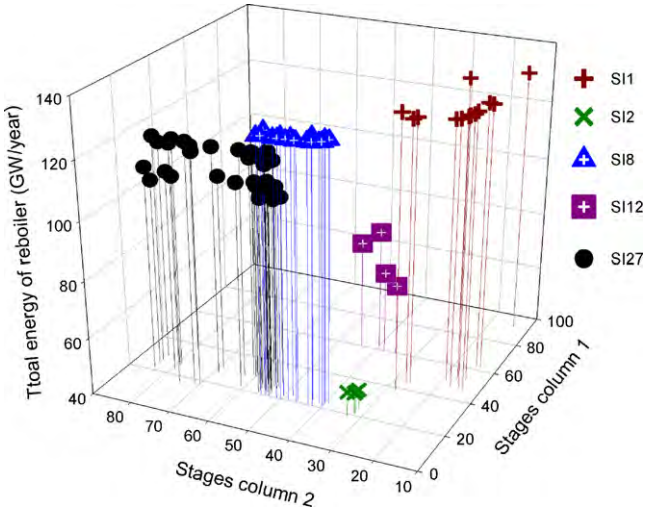

Fig. 14 – Pareto front of sequences with less than N – 1 columns, mixture M3.

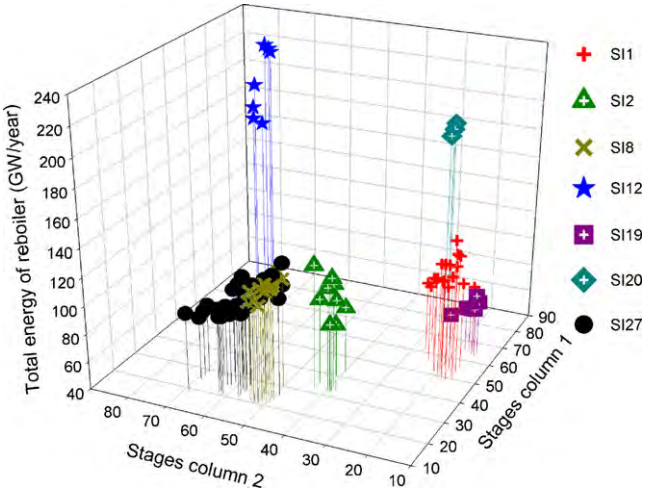

Fig. 15 – Pareto front of sequences with less than N – 1 columns, mixture M4.

For this particular case study, we see that if one of the components of the hard cut is in less proportion and the configuration of the sequence allows having the cut B/C to pass from first column to the second one, the performance of separation is better. In the sequences of this type, the minimum energy consumption can be found in those sequences without thermal coupling.

4.3. *Mixture 3*

The third mixture in our analysis is also a mixture of hydrocarbons; the light, A, intermediates, B and C, and the heavy components D are of normal chain. The feed composition contains a major concentration of the two intermediate components (90%). From the relative volatilities of the adjacent components, it is seen that the three splits of the feed components have approximately equal indexes of easy separation.

Fig. 14 showed the Pareto front of distillation sequences with less than $N-1$ columns without thermal coupling, with thermal coupling, and also with the single distillation column. Now, from Fig. 14 we observe that the lower energy consumptions are showed by sequence SI2, without thermal coupling, (only if it is compared with others intensified sequences, because for this mixture the minimum energy consumption is obtained in SC1), where the separation cut of the mixture, between column one to column two, is C/D. In the group of thermally coupled schemes, the sequence that follows in energy consumption is SI12, where we observe again that the cut C/D is performed to pass from one column to another. As was observed in mixtures M1 and M2, the lower energy consumption is observed in a sequence without thermal coupling. In these mixtures, thermally coupled sequences are not so close to the minimum energy consumption, SI2; in fact, the energy requirements are between three and seven times the energy reported in the sequences without thermal coupling. Also, the single distillation column, SI30, has a high heat duty, but the highest of all sequences for mixture M3 is SI5 (results are not displayed in Fig. 14; in particular sequence SI5 cannot obtain the target recovery, in any case analyzed).

It seems that when the configuration of the sequence allows having the cut C/D, the easiest, to pass from first column to the second one the performance of separation is better. In this case, the purification of the light and heavy components is complex due to they are in less proportion. According to the results, the best option is avoiding the separation of these two components in the same column, which implies performing the cut A/B or C/D in the first column. Considering that the easy cut, C/D, has to be realized in the first column: then the lower proportion component, D, is separated in the first column from the rest of the mixture; so, the hard separations are performed in the second column. Due to this, the sequences SI5 (SG2), SI8 (SG3), and SI28 (SG8) have the worst performance in terms of energy consumption. Nevertheless, sequence SI31, SG9 (not shown in Fig. 14) requires more than three times the energy consumed by sequence SI2, even when the number of stages of sequence SI31 is greater than the stages of scheme SI2.

From Fig. 14 we observe that the minimum energy consumption is presented in one sequence of two columns without thermal coupling, SI2; being the thermally coupled ones higher in energy consumption from this minimum value, for instance SI12. Table 4 shows thermodynamic efficiency, total annual cost and CO_2 emissions for the sequences of all groups. From this table we can observe that thermodynamic efficiency is greater in sequence SC1, followed by trains SI2, same tendency found in heat duties.

For this particular case study, we see that if two components are in very small proportion (10% or less) avoiding the sequences where these components can be purified in the same distillation column. Thereby, if the cut C/D is performed to pass from one column to another, minimum energy consumption can be found in those arrangements.

4.4. *Mixture 4*

The last mixture in our analysis is a mixture of alcohols; the light, A, and heavy, D, are isomers, while the intermediate components, B and C, are of normal chain. The feed composition contains a minor concentration of the light component (5%), and the more abundant components are an intermediate, C, and the heavy, D. The volatilities indicate that the harder cut for separation is C/D, being these components the more abundant. In this case, the most easy cut is among iso-butanol, A, and 1-pentanol, B; the harder cut includes 1-hexanol, C, and iso-heptanol, D.

Fig. 15 shows the Pareto front of distillation sequences with less than $N-1$ columns without thermal coupling, with

Table 4 – Summary of Pareto front for the intensified sequences, M3.

Intensified sequences	Group	Number sequence	Number column	Total stages	Feed stage	Side stage 1	Side stage 2	Vapor stage	Liquid stage	Reflux ratio	Diameter (m)	Total energy of reboiler (GW/year)
2 columns	SG1	DCS (conventional Direct sequence)	1	28	11	–	–	–	–	18.63	0.66	3.81
			2	24	8					3.62	0.96	6.16
			3	25	11					0.90	0.81	4.54
		SI1	1	44	31	22	–	–	–	50.66	4.30	128.07
			2	27	9	–				9.72	0.49	2.21
		SI2	1	24	10	–	–	–	–	5.46	1.94	34.00
			2	32	17	17				63.84	1.14	12.89
	SG2	SI5	1	36	6	16	–	–	–	90.00	2.03	37.86
			2	17	9	–				10.00	7.85	338.36
		SI8	1	20	11	–	–	–	–	19.80	0.72	4.73
	SG3		2	41	18	18				54.87	3.64	98.13
		SI10	1	50	38	25	–	–	–	383.13	3.18	82.84
			2	35	15	–				3.01	0.98	9.61
Coupled	SG4	SI12	1	60	19	–	–	–	–	300.00	0.97	9.44
			2	35	–	18		23	26	–	2.67	55.59
	SG8	SI27	1	27	13	–	–	–	–	44.32	0.70	–
			2	54	–	24		45	49	–	3.79	104.76
1 column	SG9	SI30	1	101	48	22	36	–	–	632.5	4.478	136.08

Table 4 (*Continued*)

Intensified sequences	Group	Number sequence	Number column	Molar flow of A	Molar flow of B	Molar flow of C	Molar flow of D	Molar flow of liquid	Molar flow of vapor	Thermodynamic efficiency	CO_2 emissions	Total annual cost
						(Mton/year)				η	(Mton/year)	(MDD/year)
2 columns		DCS (conventional Direct sequence)	1 2 3	3.09	33.63	38.62	4.54	–	–	4.50	0.30	0.70
	SG1	SI1	1 2	2.28	27.27	32.70	4.26	–	–	1.28	2.36	4.61
		SI2	1 2	2.19	27.39	32.68	4.25	–	–	3.54	0.81	1.76
	SG2	SI5	1 2	2.72	27.49	32.49	3.74	–	–	4.57	0.67	1.53
	SG3	SI8	1 2	2.41	27.10	32.69	4.26	–	–	1.61	1.85	3.69
		SI10	1 2	2.42	27.41	32.38	4.17	–	–	2.14	1.52	3.31
Coupled	SG4	SI12	1 2	3.08	33.20	38.83	4.29	51.28	–	3.13	1.05	2.44
	SG8	SI27	1 2	3.13	33.26	38.62	4.35	–	46.72	1.57	1.91	3.83
1 column	SG9	SI30	1	3.11	28.07	28.07	3.16	–	–	1.27	2.61	5.04

Table 5 – Summary of Pareto front for the intensified sequences, M4.

Intensified sequences	Group	Number sequence	Number column	Total stages	Feed stage	Side stage 1	Side stage 2	Vapor stage	Liquid stage	Reflux ratio	Diameter (m)	Total energy of reboiler (GW/year)
2 columns	SG1	DCS (conventional Direct sequence)	1	26	8	–	–	–	–	18.34	0.86	8.88
			2	51	24					3.83	1.01	11.07
			3	56	24					3.62	1.44	17.68
		SI1	1	108	44	27	–	–	–	30.91	3.03	86.11
			2	38	11	–				4.94	0.44	2.49
		SI2	1	43	16	–	–	–	–	5.74	2.13	46.47
			2	40	11	11				92.67	1.81	38.22
	SG2	SI5	1	49	15	35	–	–	–	97.38	2.17	49.00
			2	24	9	–				33.43	2.17	49.00
	SG3	SI8	1	26	14	–	–	–	–	17.09	0.84	8.23
			2	62	25	25				36.99	3.00	85.69
		SI10	1	47	32	18	–	–	–	602.23	5.77	258.22
			2	33	15	–				10.29	2.04	41.99
Coupled	SG4	SI12	1	78	24	–	–	–	–	380.86	1.40	21.38
			2	79	–	17		33	37	–	4.39	145.07
	SG6	SI19	1	65	54	21	–	34	–	150.46	2.52	62.78
			2	22	–	–		–	37	–	0.47	–
		SI20	1	77	58	42	–	11	–	438.30	4.79	177.31
			2	28	–	–		–	15	–	0.40	–
	SG8	SI27	1	29	12	–	–	–	–	33.09	0.85	–
			2	65	–	42		21	23	–	2.99	85.20
1 column	SG9	SI30	1	97	63	19	38	–	–	199.5	3.001	85.49

Table 5 (*Continued*)

Intensified sequences	Group	Number sequence	Number column	Molar flow of A	Molar flow of B	Molar flow of C (Mton/year)	Molar flow of D (Mton/year)	Molar flow of liquid	Molar flow of vapor	Thermodynamic efficiency η	CO_2 emissions (Mton/year)	Total annual cost (MDD/year)
2 columns	SG1	DCS (conventional Direct sequence)	1 2 3	3.11	18.59	34.26	29.23	–	–	2.00	2.39	1.67
		SI1	1 2	3.02	18.32	34.32	29.65	–	–	3.28	2.22	2.78
		SI2	1 2	2.86	18.30	34.60	29.61	–	–	2.63	2.77	3.38
	SG2	SI5	1 2	3.44	18.67	33.20	29.83	–	–	1.86	3.01	3.81
	SG3	SI8	1 2	3.07	18.33	34.32	29.57	–	–	2.14	3.03	3.76
		SI10	1 2	3.06	18.65	28.84	29.15	–	–	0.68	9.52	10.89
Coupled	SG4	SI12	1 2	3.12	74.14	4.28	25.70	19.03	–	1.41	5.23	6.43
	SG6	SI19	1 2	3.07	18.49	34.38	25.77	–	35.31	2.52	1.81	2.59
		SI20	1 2	3.14	18.46	34.19	25.89	27.90	–	0.89	5.08	6.71
	SG8	SI27	1 2	2.45	15.01	28.80	21.92	–	66.76	2.32	2.77	3.40
1 column	SG9	SI30	1	3.04	15.44	24.94	18.99	–	–	2.36	2.78	3.55

thermal coupling, and also with the single distillation column. Now, from Fig. 15 we observe that the lower energy consumptions are showed by sequence SI20, with thermal coupling, where the separation cut of the mixture is performed between the columns is C/D. In the group of schemes without thermal coupling, the sequence that follows in energy consumption is SI2, where, again, we observe that the cut C/D is performed to pass from one column to another. In contrast with the behavior observed in the previous mixtures, the lower energy consumption is observed in a sequence with thermal coupling. In this mixture, a significant difference in energy consumption is observed between sequence with and without thermally coupled sequences. Also, the single distillation column, SI30, has a heat duty higher than the minimum value, but the highest of all sequences for mixture M4 is SI10, Table 5.

It seems that when the configuration of the sequence allows having the cut C/D to pass from first column to the second one the performance of separation is better. In this case, the purification of one intermediate and heavy component is hard, but they are in major proportion in the feed stream. Also, it is important to avoid making the cut considering the split A/B; in this case, the difference in relative volatilities is big but the light component is present in a minor proportion (less than 10%). Due to this, the sequences SI12 (SG4), SI10 (SG3), and SI21 (SG6) have the worst performance in terms of energy consumption. Nevertheless, sequence SI30 (SG9) has not the higher energy consumption for this mixture.

From Fig. 15 we observe that the minimal energy consumption is presented in one sequence of two columns with thermal coupling, SI20; being the schemes without thermal coupling higher in energy consumption from this minimum value, for instance SI10. The Table 5 shows thermodynamic efficiency, total annual cost and CO_2 emissions for the sequences of all groups. From this table we can observe that thermodynamic efficiency is greater in sequence SI1, followed by trains SI2; without doubt, a different tendency is found in heat duties, being the minimal energy presented in sequence SC1. As consequence, in sequence SI20 the CO_2 emissions are lower.

For this particular case study, we see that if the light component presents the lowest composition, the minimum energy consumption can be found if the sequence allows having the cut C/D to pass from first column to the second one the performance of separation, in those with thermal coupling.

4.5. *Other observations for this case study*

In the analysis presented before, some recommendations about the best scheme to separate a mixture with certain feed composition and relative volatilities are given. In this section, some other findings related to the sequences that are not promising are given, in order to be applied in other study cases.

In general, the intensified sequences of two columns that are linked between them through side streams are not feasible for the separation of quaternary mixtures; this due the purities and recoveries are not reached. We found that the feasibility of an intensified scheme it depends on the feed composition, while the relative volatilities influence the energy consumption of the feasible scheme. Now, the feasibility of the scheme can be verified using distillation lines, since the columns analyzed in this study are staged. In order to verify this observation, we calculate distillation lines that pass through a distillate composition for instance; then, we expect that in this line the compositions of side and bottom streams were

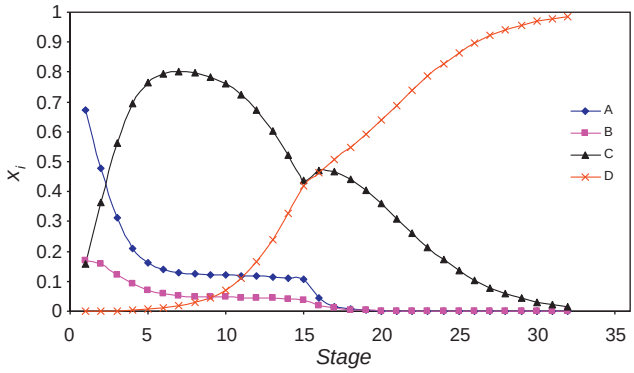

Fig. 16 – Composition profiles in liquid phase of the first column of sequence SI2, mixture M1.

located. The resulting line show that required compositions cannot be located in this line. Then, these schemes are not feasible to separate the mixtures under consideration. In contrast, the schemes of two columns with no side streams show a good behavior for the most of the cases analyzed, presenting lower energy consumptions and a reduced number of stages. These sequences are the best example of a desirable intensified process: reduction in capital costs (number of stages) with considerable heat duties, this in comparison with conventional sequences ($N-1$ columns).

An unexpected behavior is that the use of intensified sequences of two columns that are thermally coupled show higher energy consumptions, just below of the single distillation columns with two side streams. From the obtained results it is clear that the energy consumption in these sequences is higher than the requirements in schemes without thermal coupling. Figs. 16 and 17 show the composition profiles of the first column of sequences SI2 and SI12 for mixture M1. From these figures it is clear that component C is separated with a high purity in the first column of sequence SI12, with thermal coupling, but this component is not extracted until the second column. On the other hand, the enrichment of the same component in the first column of sequence SI2 is minor. The energy used to over purify C in the first column of the sequence with thermal coupling is the main reason of the increasing in the energy consumption in thermal coupled sequences. It seems that the principal feature that enables reducing energy in sequences with $N-1$ columns with thermal coupling is the one that causes increasing in energy consumption when the thermally coupled sequences contains less than $N-1$ columns.

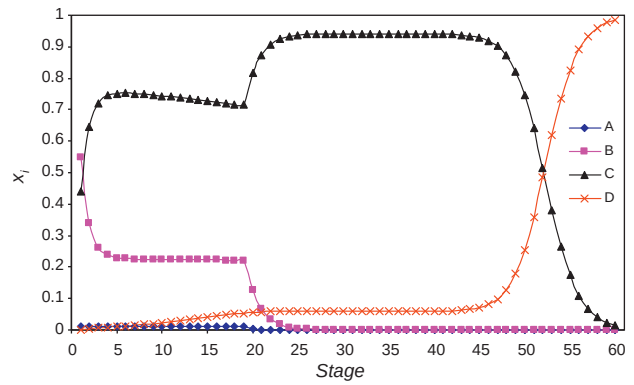

Fig. 17 – Composition profiles in liquid phase of the first column of sequence SI12, mixture M1.

Finally, the intensified sequences with just one distillation column present, in general, the higher energy consumptions of all studied sequences. In these columns, the reflux ratio is very high since the components obtained in the side streams require high purities and recoveries. Also, an elevated number of stages is required.

5. Conclusions

A design methodology for generating optimal designs of intensified sequences with less than $N-1$ distillation columns has been presented. As design tool we used a multiobjective genetic algorithm with constraints, which is coupled to Aspen Plus in order to evaluate the objective and constraints functions.

Results show that it is possible to separate mixture in these sequences with less than $N-1$ columns, with and without thermal coupling. The best configuration it depends on the nature of the mixture, feed composition, purities and recoveries required. For the mixtures that we analyze, we found that sequences of less than $N-1$ columns without thermal coupling present the minor energy consumptions of all studied sequences. This result was quite unexpected, considering that in sequences with $N-1$ columns the thermally coupled ones present minor energy consumptions.

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Appendix A.

The thermodynamic efficiencies of optimized designs of the TCDS schemes can be computed using the laws of the thermodynamic. We used for this task the equations reported in the textbook by Seader and Henley (2006). The equations are:

First Law of the thermodynamic:

$$\sum_{\text{out of system}} (nh + Q + W_s) - \sum_{\text{into system}} (nh + Q + W_s) = 0 \quad \text{(A.1)}$$

Second Law of the thermodynamic:

$$\sum_{\text{out of system}} (ns + Q/T_s) - \sum_{\text{into system}} (ns + Q/T_s) = \Delta S_{irr} \quad \text{(A.2)}$$

Exergy balance:

$$\sum_{\text{into system}} \left[nb + Q\left(1 - \frac{T_0}{T_S}\right) + W_s\right] - \sum_{\text{out of system}} \left[nb + Q\left(1 - \frac{T_0}{T_S}\right) + W_s\right] = LW \quad \text{(A.3)}$$

Minimum work of separation:

$$W_{\min} = \sum_{\text{out of system}} nb - \sum_{\text{into system}} nb \quad \text{(A.4)}$$

Second law efficiency where η is the thermodynamic efficiency:

$$\eta = \frac{W_{\min}}{LW + W_{\min}} \quad \text{(A.5)}$$

The exergy function:

$$b = h - T_0 s \quad \text{(A.6)}$$

The lost work in the system:

$$LW = T_0 \Delta S_{irr} \quad \text{(A.7)}$$

The thermodynamic properties like enthalpies and entropies of the streams of the distillation sequences were evaluated through the use of the simulator of processes Aspen One v12.1™.

Appendix B.

The calculation of CO_2 Emissions used framework Gadalla et al. (2005). Fuel is combusted when mixed with air, producing Dioxide of Carbon (CO_2) according to following stoichiometric equation:

$$C_xH_y + \left(x + \frac{y}{4}\right) O_2 \rightarrow xCO_2 + \frac{y}{2}H_2O \quad \text{(B.1)}$$

where x and y denote the number of carbon, C, and hydrogen, H, atoms, respectively, present in the fuel compositions, and where complete oxidation of carbon is assumed. The CO_2 emissions, $[CO_2]_{Emiss}$ (kg/s), are related to the amount of fuel burnt, Q_{Fuel} (kW), in a heating device as follows (Gadalla et al., 2005):

$$[CO_2]_{Emiss} = \left(\frac{Q_{Fuel}}{NHV}\right)\left(\frac{C\%}{100}\right)\alpha \quad \text{(B.2)}$$

where α (=3.67) is the ratio of molar masses of CO_2 and C, while NHV (kJ/kg) represents the net heating value of a fuel with a carbon content of C%. This is Equation B.1 shows that both: the fuel used and the heating device affect the amount of CO_2 produced.

The same theoretical flame temperature of 1800 °C may be still used. The stack temperature of 160 °C is also used in the calculations. The amount of fuel burnt can be calculated from (Gadalla et al., 2005):

$$Q_{Fuel} = \frac{Q_{Proc}}{\lambda_{Proc}}(h_{Proc} - 419)\ \frac{T_{FTB} - T_0}{T_{FTB} - T_{Stack}} \quad \text{(B.3)}$$

where λ_{Proc} (kJ/kg) and h_{Proc} (kJ/kg) are the latent heat and enthalpy of steam delivered to the process, respectively, while T_{FTB} (°C) is the flame temperature of the boiler flue gases.

Appendix C.

The costing of distillation column (carbon steel construction) was estimated by the cost equations showed in Turton et al. (2004) which are updated with the CEPCI (Chemical Engineering Process Cost Index, in this case was 575.4).

The capital cost (purchase plus installation cost) is annualized over a period that is often referred to as plant life time, in this case is 5 years, considered 8400 h/y of operation:

$$\text{Annual capital cost} = \frac{\text{Capital cost}}{\text{Plant life time}} \quad \text{(C.1)}$$

$$\text{Total Annual Cost (TAC)} = \text{Annual operating cost} + \text{Annual capital cost} \quad \text{(C.2)}$$

The total column cost is the sum of the installed cost of column shell and the installed cost of column trays. The sizing and costing of heat exchangers were calculated. The cost of heat exchangers can be correlated as a function of the surface area assuming shell and tube, floating head, and carbon steel construction.

For a given number of theoretical trays, Aspen Plus simulator calculates column diameter and height (for 24 in tray spacing) after converging for selected valve tray column with 2 in weir height. Valve trays of Sieve type are considered.

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