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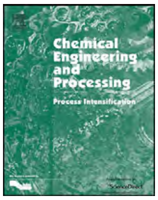

Hybrid Distillation/Melt Crystallization Process Using Thermally Coupled Arrangements: Optimization with evolutive algorithms

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ABSTRACT

Innovative hybrid processes offer significant cost savings, particularly for azeotropic or close-boiling mixtures. Hybrid separation processes are characterized by the combination of two or more different unit operations, which contribute to the separation task by different physical separation principles. Despite of the inherent advantages of hybrid separation processes, they are not systematically exploited in industrial applications due to the complexity of the design and optimization of these highly integrated processes. In this work we study a hybrid distillation/melt crystallization process, using conventional and thermally coupled distillation sequences. The design and optimization were carried out using, as a design tool, a multi-objective genetic algorithm with restrictions coupled with the process simulator Aspen Plus^{TM}, for the evaluation of the objective function. The results show that this hybrid configuration with thermally coupled arrangements is a feasible option in terms of energy savings, capital investment and control properties.

1. Introduction

The chemical industry is the largest consumer of energy of the manufacturing industries; it represents 6% of all domestic energy use, and 24% of the total U.S. manufacturing energy use. Petroleum refining is the second largest consumer of energy, with a contribution of approximately 10% to the total U.S. manufacturing energy use. Separations assess for approximately the 60% of the in-plant energy usage for these two industries. Distillation operations account for near to 95% of the total separation energy used in the refining and chemical processing industries with about of 40,000 distillation columns operating in over 200 different processes [1]. This high usage rate is primarily due to distillation's flexibility, low capital investment relative to other separations technologies and low operational risk. Unfortunately, the energy efficiency of a commercial distillation column is low, with a second-law efficiency of less than 10% being typical [2]. This is reflected on high quantities of thermal energy required to achieve the desired purification. Thus, there is a major research opportunity area on the development of improvements or replacements for distillation to achieve significant energy savings, due to the large sunk capital investment in existing plants, the slow rate of plant replacements, and the diverse numbers of applications where distillation is utilized. Such enhancements to the distillation operation may have particular importance in separations with low relative volatilities, or those that operate at cryogenic or very high temperatures.

Opportunities for improving equipment in existing distillation systems include divided wall columns, improved packing designs, heat integrated distillation, and improving mass transfer efficiencies; these options have already been studied [3–6]. Independent technical reviews of industrial experiences to date and pilot-plant demonstrations are needed for wide-spread implementation of divided wall columns and heat integrated distillation systems. The debottlenecking opportunities described above can not be implemented generically across the industry; they will be process flowsheet specific. Hybrid and improved equipment systems are already being used to a limited extent within the industry, and there are improved mass separating agents and process equipment on the market today that are not being used extensively by the industry for debottlenecking plants. This is in part due to lack of tools to evaluate their performance for specific applications [1].

In recent years, there has been an increasing interest on hybrid process in chemical engineering (see [7–11], among others). Because of inherent drawbacks of conventional processes, enhancement of such processes represents an important field of opportunity to the development of technological advances.

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Nomenclature

F	feed flowrate
F_j	Molar flowrate of the interconnection flow
$\overline{H}$	enthalpy of vapor
$\overline{h}$	enthalpy of liquid
ΔH_{m}	melting enthalpy
ΔH_{v}	vaporization enthalpy
K	equilibrium constant
L	liquid flowrate
M	moles of liquid retained
N_i	number of stages for the column i
N_j	location of the interconnection flow j
N_{s}	location of the side stream
$N_{\mathrm{F,i}}$	location of the feed stage
Q	total energy requirements
Q_i	energy requirements for the column i
R	reflux ratio
T_{b}	boiling temperature
T_{m}	melting temperature
t	time
U	liquid side stream
$\overline{U}$	internal energy of liquid retained
u	manipulated input
V	vapor flowrate
W	vapor side stream
X	mol fraction in liquid phase
$\bar{x}_k$	vector of required purities
x_{f}	composition in the feed
$\bar{y}_k$	vector of obtained purities
Y	mol fraction in vapor phase

Subscripts

i	component
j	stage

Greek symbols

α_{ij}	relative volatility
γ	condition number
σ^*	minimum singular value

Therefore, the focus of separation and purification studies is gradually being shifted from unit operations towards hybrid processes. One example of a hybrid processes involves the combination of the well-known crystallization operation with the ease of phase separation typical of distillation. They are applicable to solid–liquid–vapor three-phase equilibrium, and it may result useful for the separation of mixtures with pairs of components with relative volatilities close to 1. In such systems, the liquid, enriched in the impurities that have not passed into the crystals, vaporizes at a low pressure, near the triple-point pressure of the main component. Therefore, combining all these processes in one installation is expected to raise the separation efficiency without requiring any considerable extra expenses [9]. Another important hybrid process corresponds to the combination of distillation and melt crystallization for separation of close-boiling isomer mixtures. The hybrid distillation/melt crystallization process combines advantages of the distillation and the melt crystallization, in which very high separation factors per stage can be reached. Simultaneously, the combination of distillation and crystallization overcomes the shortcomings of the individual unit operations, i.e. high energy requirements at small separation factors, and limitation of yield by eutectics, respectively. Several hybrid processes for the separation of terphenyl, xylene, dichlorobenzene and diphenylmethane diisocyanate isomers are reported in literature. Berry and Ng [7] used simplified models, based on constant separation factors, for the synthesis of hybrid distillation/melt crystallization processes and proposed guidelines for flowsheet selection. Wallert et al. [12] also applied shortcut methods for the synthesis and evaluation of a hybrid distillation/melt crystallization processes. Franke et al. [10] proposed a three-step design method for hybrid distillation/melt hybrid processes. In a first step different sequences are generated by heuristic rules. These sequences are evaluated in a second step by shortcut methods on the basis of energy requirements, in order to identify the most promising alternatives. In the third and last step a reduced number of promising sequences is rigorously optimized by MINLP methods, and the best sequence on the basis of total annualized costs is chosen. It should be noted that the creation of a superstructure is not a trivial task, especially for hybrid separation processes. Marquardt et al. [13] reviewed an optimization-based framework composed of shortcut and rigorous design steps for the robust and efficient synthesis of hybrid distillation/melt hybrid processes. A multitude of hybrid processes composed of distillation and melt crystallization units are evaluated with powerful shortcut models. A selection of promising process variants is subsequently rigorously optimized by an economic objective function and discrete-continuous optimization techniques. It is shown that the design of the cost-optimal hybrid process within the systematic synthesis framework can be accomplished with robustness and efficiency. However, the main drawback of a mathematical programming method is that they cannot guarantee to find the global optimum if non-convex equations are present, and, also, in spite of their high computational effort they often fail to solve large scale process engineering problems with highly non-lineal and mixed-integer models [14,15].

On the other hand, the presence of recycle streams for complex distillation schemes has generated the idea that control problems might be expected during the operation those arrangements with respect to behavior of conventional distillation configurations. Understanding control properties of process with distillation columns with thermal couplings is an issue of extreme importance since designs with economic incentives often conflict with their operational characteristics. However, recent publications report considerable progress in the identification of suitable control variables and control strategies for some configurations with thermal coupling [16–19].

In this work, the design and optimization of a hybrid distillation/melt crystallization process with conventional and thermally coupled distillation sequences is presented. We select the use of distillation configurations with thermally coupling due to the energy savings, total annual cost reduction and good dynamic behavior of those arrangements in comparison with properties of conventional distillation structures (see [5,20–22] among others). The design and optimization were carried out using a multi-objective genetic algorithm with restrictions handling, coupled with the process simulator Aspen Plus$^{\mathrm{TM}}$, for the evaluation of the objective function, ensuring that all results considered full use of the equations of the models contained in the Aspen Plus simulator. Rudolph [23] proved that genetic algorithms, a kind of evolutive algorithms, converge to the global optimum. To the best of our knowledge, multiobjective stochastic methods have not been reported for optimal design of a hybrid distillation/melt crystallization process with conventional and non-conventional distillation arrangements. For each analyzed system a set of optimal designs, called Pareto front, is obtained. The results show that the hybrid configuration with thermally coupled arrangements is a feasible option, presenting advantages over configurations with conventional distillation systems in terms of energy savings (and, consequently, reductions in greenhouse gas emissions), capital investment and control properties.

2. Optimization strategy

In the last years, stochastic methods have been successfully applied to process engineering optimization problems [24–30]. Stochastic optimization methods are playing an important role because they are generally robust numerical tools, which present a reasonable computational effort in the optimization of multivariable functions; they are also applicable to unknown structure problems, requiring only calculations of the objective function, and can be used with all models without problem reformulation. Moreover, a major advantage of genetic algorithms over other stochastic techniques is the availability of several multiobjective techniques such as VEGA, MOGA, NSGA, Niche Pareto GA, and NSGA-II [30]. These stochastic methods are very useful for the reliable design and optimization of chemical processes, where several decision variables are involved. To the best of our knowledge, multiobjective stochastic methods have not been reported for design and optimization of the hybrid process with rigorous models. The detailed design of a hybrid separation process is a challenging task because of many degrees of freedom involved [10]. In order to optimize the hybrid distillation/melt crystallization process, using conventional and thermally coupled distillation sequences, the multiobjective genetic algorithm with constraints coupled to Aspen ONE Aspen Plus developed by Gutiérrez-Antonio and Briones-Ramírez [30] has been used. The algorithm manages the multiobjective optimization using the NSGA-II [31], and the constraints are handled taking as a basis the concept of non-dominance proposed by Coello-Coello [32]. A brief description of the process is presented below. The entire population is divided into sub-populations using, as criterion, the total number of satisfied constraints. Thus, the best individuals of the generation are those that satisfied the n constraints, and they are followed by the individuals only satisfying $n-1$, and so. Within each sub-population, individuals are ranked using the NSGA-II, but considering now as other objective function to minimize the degree of unsatisfied constraints. Next, dominance calculation of each subgroup is carried out as follows:

$$\text{dominance}\{Q_i, N_i, \min[0, (\vec{x}_k - \vec{y}_k)]\} \tag{1}$$

In terms of multiobjective optimization, when a minimization takes place and the algorithm reaches a point where there is no feasible vector that can decrease the value of one objective without simultaneously increasing the value of another objective, it is said that point in the search space is the Pareto optimum. By definition we can say that one point $\bar{z}^* \in \Im$ is a Pareto optimum if for each $\bar{z} \in \Im$:

$$\underset{n \in I}{\wedge} (f_n(\bar{z}) \geq f_n(\bar{z}^*)) \tag{2}$$

or at least there is some $n \in I$, where I represents the set of objective functions to optimize, that:

$$f_n(\bar{z}) > f_n(\bar{z}^*) \tag{3}$$

Then, we define that $\vec{z}$ dominates $\vec{w}$ when $f(\vec{z}) < f(\vec{w})$, if $W \in \Im$ and $\vec{w} \in W$ if none $\vec{z} \in W$ dominates $\vec{w}$, we say that $\vec{w}$ is not dominated with respect to W. As established by Mezura-Montes [33], the set of solutions which are not dominated and optima of Pareto integrates the Pareto front. For unit operations, the Pareto front represents all optimal designs, from minimum energy requirements to minimal size of equipment. The adequate design shall be chosen by selection of a point along the Pareto front.

In the optimization problem of the hybrid distillation/melt crystallization process, the simultaneous minimization of the energy requirements and the size of the equipments involved in the sequence are considered: for the case of the distillation columns is estimated diameter and height of the column and the condenser and reboiler areas. In the case of the crystallizer is estimated the volume of the equipment. The link to Aspen Plus allows have optimal designs using the full model of the equipments with all equations considered in the simulator; however, the 95% of the total time of the optimization procedure is employed in performing these simulations. To reduce this time, artificial neuronal networks, ANN, are used to speed up the multiobjective genetic algorithm with constraints [30]. Neuronal networks generate approximate functions for objectives and constraints. The approximated and complete objectives and constraints functions are switched during the optimization; in order to improve their prediction, neural networks are retraining when the complete functions are used. Fig. 1 shows a block diagram for the evolutionary strategy coupled to the artificial neuronal networks.

For the optimization hybrid system, we used 800 individuals and 40 generations as parameters of the multiobjective genetic algorithm, with a frequency parameter was 5. These parameters were obtained through a tuning process, where several runs of the algorithm were performed with different numbers of individuals and generations.

3. Analyzed configurations

Four different hybrid configurations have been analyzed: an arrangement consisting on melt crystallization with conventional distillation (C-DSI-C, Fig. 2); an arrangement of a melt crystallization system with a thermally coupled indirect sequence (C-TCDS-C, Fig. 3); an arrangement of melt crystallization with a modified thermally coupled indirect sequence (C-MTCDS-C, Fig. 4); and an arrangement consisting on melt crystallization with a Petlyuk column (C-PC-C, Fig. 5). For all the systems studied, the use of the cheapest refrigerant must be ensured. The hybrid configurations proposed in this study have been obtained following the rules of Berry and Ng [7] for the synthesis of hybrid sequences. The synthesis procedure proposed by Berry and Ng [7] allows obtaining hybrid configurations with low values of total annual cost.

For the C-DSI-C arrangement, the optimization problem has been formulated as:

$$\begin{aligned} &\min(Q_i, N_i) = f(R, N_i, N_{\mathrm{F},i}) \\ &\text{s.t.} \\ &\vec{y}_k \geq \vec{x}_k \end{aligned} \tag{4}$$

where Q_i and N_i are the heat duty and number of stages of the column i; the objectives for the optimization of this arrangement are four: two for the heat duty and two for the number of stages. The variables involved in the optimization of the hybrid sequences are now presented. R is the reflux ratio, N_i is the total number of stages of the column i, $N_{\mathrm{F},i}$ is the location of feed stage, $\vec{x}_k$ and $\vec{y}_k$ are vectors of required and obtained purities or recoveries for the k components.

In the case of the C-TCDS-C configuration, the minimization problem has been formulated as:

$$\begin{aligned} &Min(Q_i, N_i) = f(R, N_i, N_j, N_{F,i}, F_j) \\ &st \\ &\vec{y}_k \geq \vec{x}_k \end{aligned} \tag{5}$$

where Q_i and N_i are the heat duty and number of stages of the column i; the objectives for the optimization of this arrangement are four: two for the heat duty and two for the number of stages. The variables involved in the optimization of the hybrid sequences are now presented. R is the reflux ratio, N_i is the total number of stages of the column i, $N_{\mathrm{F},i}$ is the location of feed stage, N_j is the location of the interconnection flow j, N_{F} is the location of feed stage, F_j is the flowrate of the interconnection flow j, $\vec{x}_k$ and $\vec{y}_k$ are vectors of required and obtained purities or recoveries for the k components.

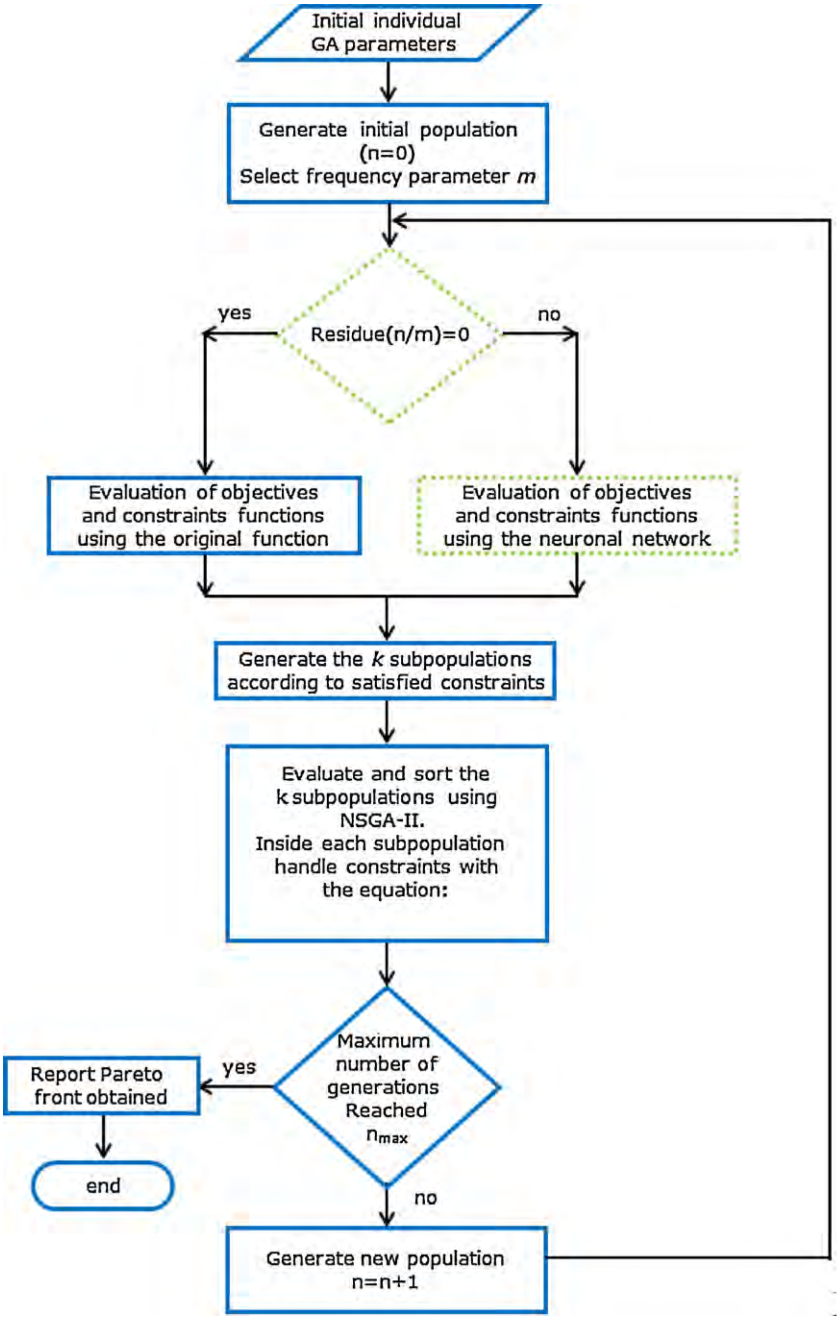


Fig. 1. Optimization strategy.

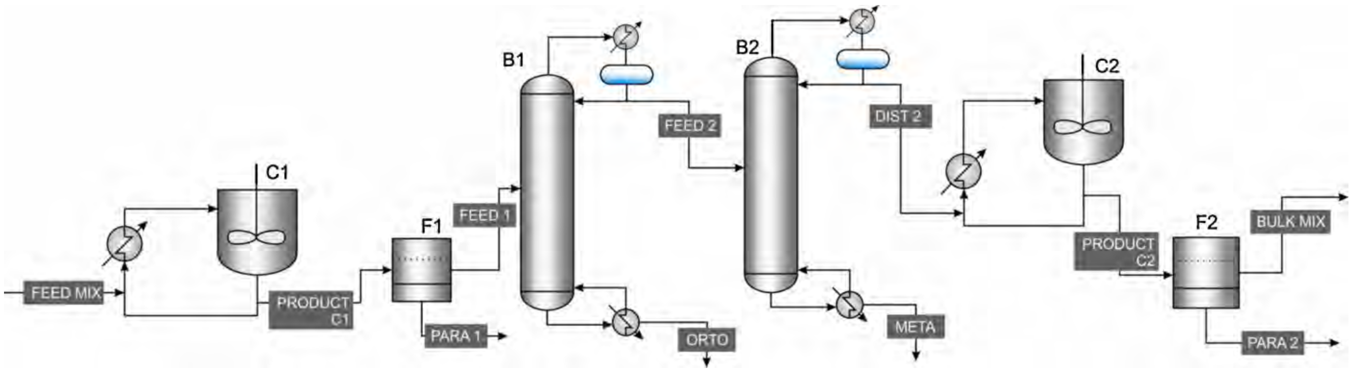


Fig. 2. Hybrid distillation/melt crystallization processes using conventional distillation arrangement (C-DSI-C).

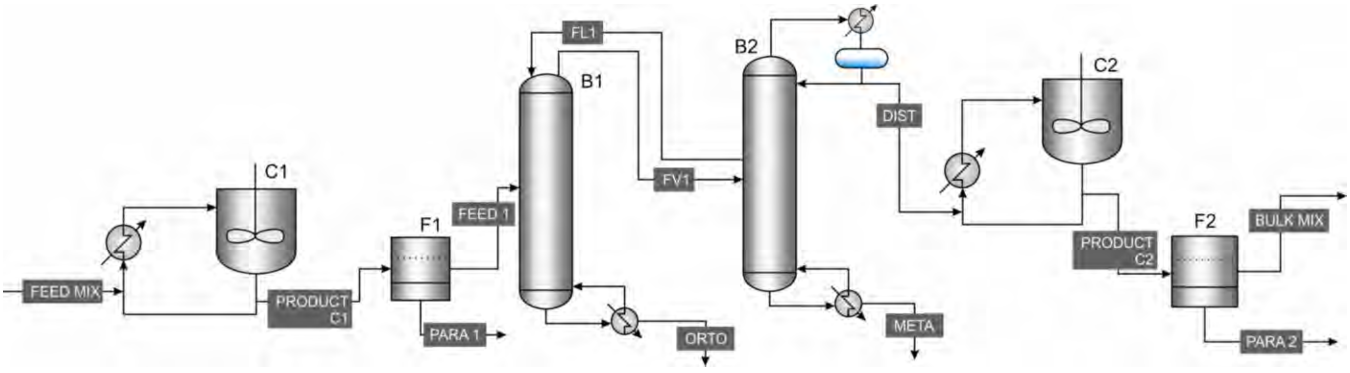


Fig. 3. Hybrid distillation/melt crystallization processes using an indirect thermally coupled distillation sequence (C-TCDS-C).

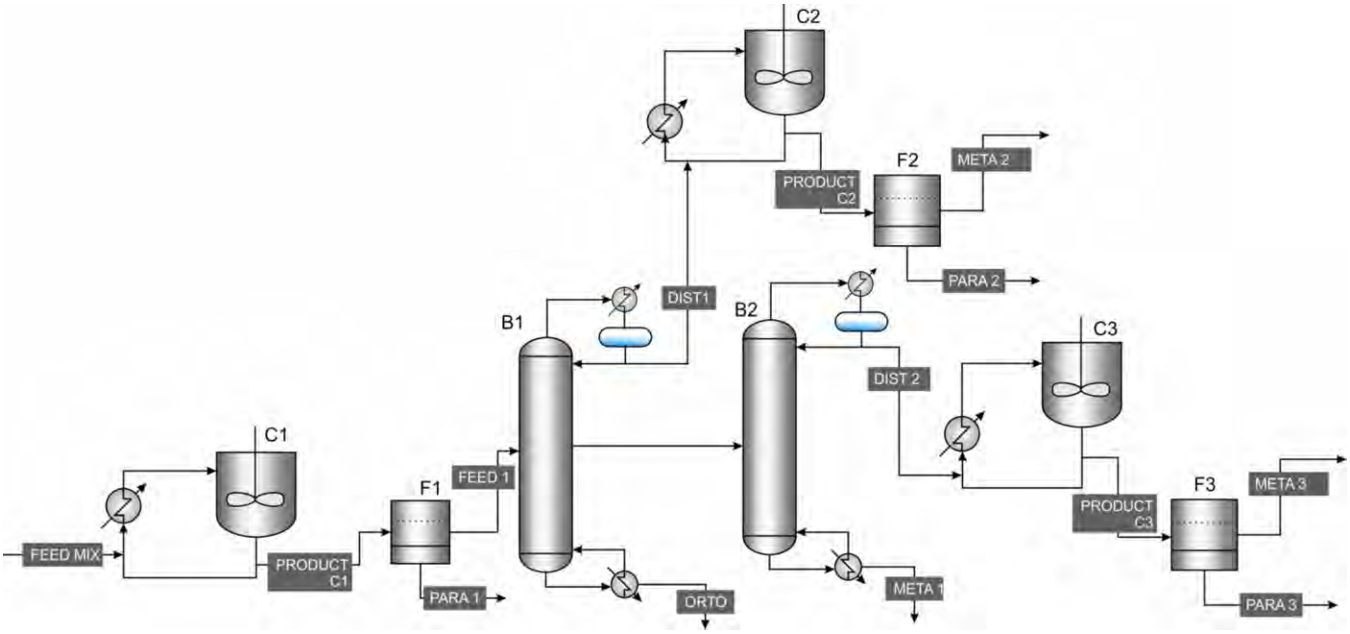


Fig. 4. Hybrid distillation/melt crystallization processes using a modified arrangement from the indirect thermally coupled distillation sequence (C-MTCDS-C).

For the C-MTCDS-C arrangement, the optimization problem has been formulated as:

$$\min(Q_i, N_i) = f(R, N_i, N_s, N_{F,i}) \quad \text{s.t.} \quad \vec{y}_k \geq \vec{x}_k \tag{6}$$

where Q_i and N_i are the heat duty and number of stages of the column i; the objectives for the optimization of this arrangement are four: two for the heat duty and two for the number of stages. The variables involved in the optimization of the hybrid sequences are now presented. R is the reflux ratio, N_i is the total number of stages of the column i, $N_{F,i}$ is the location of feed stage, N_j is the location of the interconnection flow j, N_s is the location of the side stream, N_F is

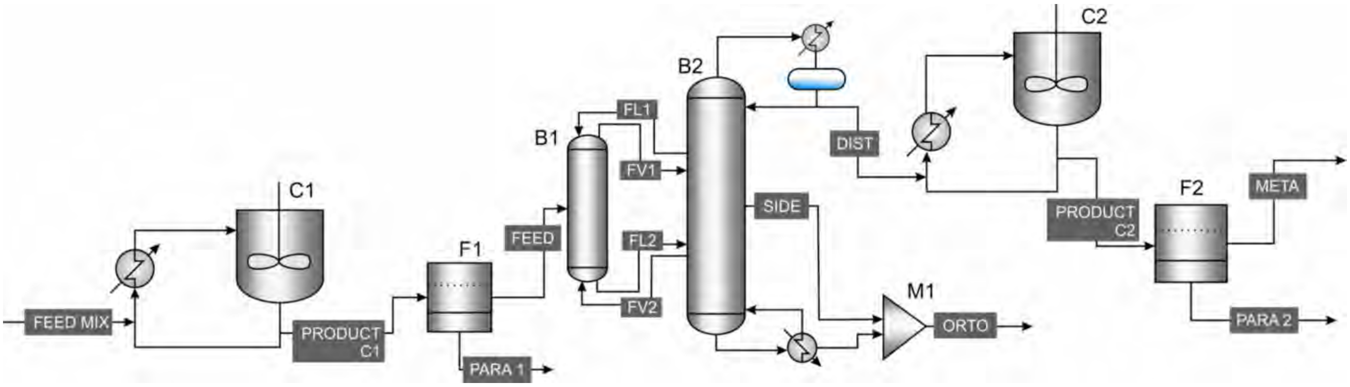


Fig. 5. Hybrid distillation/melt crystallization processes using a Petlyuk column (C-PC-C).

the location of feed stage, F_j is the flowrate of the interconnection flow j, $\vec{x}_k$ and $\vec{y}_k$ are vectors of required and obtained purities or recoveries for the k components.

Finally, for the C-PC-C configuration, this minimization problem is formulated as:

$$\begin{aligned} &\min(Q, N_i) = f(R, N_i, N_j, N_s, N_{F,i}, F_j) \\ &\text{s.t.} \\ &\vec{y}_k \geq \vec{x}_k \end{aligned} \quad (7)$$

where Q_i and N_i are the heat duty and number of stages of the column i; the objectives for the optimization of this arrangement are three: one for the heat duty and two for the number of stages. The variables involved in the optimization of the hybrid sequences are now presented. R is the reflux ratio, N_i is the total number of stages of the column i, $N_{F,i}$ is the location of feed stage, N_j is the location of the interconnection flow j, N_s is the location of the side stream, N_F is the location of feed stage, F_j is the flowrate of the interconnection flow j, $\vec{x}_k$ and $\vec{y}_k$ are vectors of required and obtained purities or recoveries for the k components. It should be noted that the total annual cost and CO_2 emissions are calculated from the design variables resulting from optimal schemes, i.e., are not objective optimization. Also, in the calculation of the total annual costs data from distillation columns and crystallizers are considered.

For the case of distillation columns, the rigorous model used in Aspen ONE Aspen Plus, is based on the following set of equation (with all symbols defined in the Nomenclature section).

Total mass balance:

$$\frac{dM_j}{dt} = L_{j-1} + V_{j+1} + F_j^L + F_j^V - (L_j + U_j) - (V_j + W_j) \quad (8)$$

Component mass balance:

$$\begin{aligned} \frac{d(M_j X_{i,j})}{dt} &= L_{j-1} X_{i,j-1} + V_{j+1} Y_{i,j+1} + F_j^L Z_{i,j}^L + F_j^V Z_{i,j}^V \\ &\quad - (L_j + U_j) X_{i,j} - (V_j + W_j) Y_{i,j} \end{aligned} \quad (9)$$

Energy balance:

$$\begin{aligned} \frac{d(M_j \overline{U}_{i,j})}{dt} &= L_{j-1} \overline{h}_{j-1} + V_{j+1} \overline{H}_{j+1} + F_j^L \overline{h}_j^L + F_j^V \overline{H}_j^V - (L_j + U_j) \overline{h}_j \\ &\quad - (V_j + W_j) \overline{H}_j + Q_j \end{aligned} \quad (10)$$

Equilibrium relationships:

$$Y_{i,j} = K_{i,j} X_{i,j} \quad (11)$$

Summation constraints:

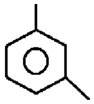
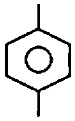
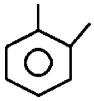
$$\sum_{i=1}^{NC} K_{i,j} X_{i,j} - 1.0 = 0 \quad (12)$$

In the case of the crystallizers we use the model proposed in Aspen ONE Aspen Plus. This model involves only the material and energy balances, considering the solubility as a parameter to determine the final degree of crystallization. Xilenes solubility data has been taken of Porter and Johnson [34]. The speed of crystal growth is not considered and the crystallization process has considered continuous operation.

4. Control properties: singular value decomposition

To complement this study, we analyze the control properties of the conventional and thermally coupled hybrid distillation/melt crystallization process based in a study of singular value decomposition technique. For more details about SVD see Klema and Laub [35], for example. First, open loop dynamic responses to changes in the manipulated variables around the assumed operating point were obtained. An important point to note is that in Aspen Plus models are also available in Aspen Dynamics making it possible to rigorously solve the dynamic model for studies of controllability. The responses were obtained through the use of Aspen Dynamics, and adjusted to proper transfer functions (i.e., first order, second order, etc., depending on the performance of the dynamic response). Transfer function matrices (G) were then collected for each case, and they were subjected to SVD:

Table 1
Composition in the feed (x_f) and separation factors ($\alpha_{i,j}$) for the isomer mixture.

Component	Formula	x_f	α_{ij}
M-Xylene		0.01	1.25
P-Xylene		0.66	1.09
O-Xylene		0.33	1.00

One definition of SVD is:

$$G = V \Sigma W^H \quad (13)$$

Here, G is the matrix target for SVD analysis and Σ is a diagonal matrix which consists of the singular values of G. The singular values of the open-loop frequency function matrix of a process at a given frequency are the gains of the process at this frequency in the directions of the corresponding input singular vectors (as the input singular vectors form a basis in the input space, the gain can be calculated in every direction). These gains play an important role when performing controllability analysis of a process, and for a complex analysis they must be evaluated at various frequencies. The Morari Resiliency Index (MRI) is the smallest singular value (σ^*) of the process open-loop frequency function matrix. The larger its value, the more controllable the process is. If it is zero, this means that there is an input direction where the gain is zero and the matrix is not invertible. Condition number (γ) is the ratio of the largest and smallest singular values of the process open-loop frequency function matrix. If it is large, then the matrix has strong directionality, which means that the gains vary strongly depending on input directions. Such a matrix is said to be ill-conditioned. A large γ means that the system is sensitive to input and model uncertainty and therefore the process is less controllable. Systems with higher σ^* values and lower γ are expected to show the best dynamic performance under feedback control [36].

5. Case of study

The design is explained with an industrial process example. The task is to separate a mixture of ortho, meta, and para-xylene isomers [37,10], with a molar composition of 1% for the low boiling meta component (M), 66% of the intermediate boiling para component (P), and 33% of the high boiling ortho component (O), as proposed by Franke et al. [10]. For each component, a purity of 99% is demanded. For the considered example, distillation alone is feasible, but economically not attractive because of the low separation factors (Table 1). Note that the separation of components P and O is especially difficult using conventional distillation, but the combination with melt crystallization offers the advantage of obtaining

Table 2
Physical properties for the case of study.

Component	Formula	Molecular weight	T_m [°C]	ΔH_m [kJ/mol]	T_b [°C]	ΔH_v [kJ/mol]
P-Xylene	C8H10	106.16	13.26	17.110	138.37	36.07
M-Xylene	C8H10	106.16	−47.87	11.569	139.12	36.40
O-Xylene	C8H10	106.16	−25.18	13.598	144.41	36.82

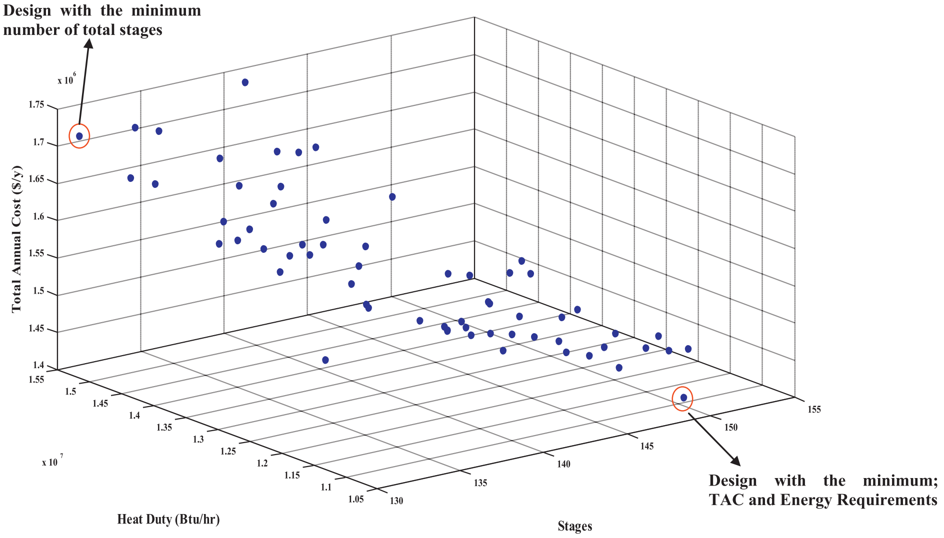


Fig. 6. Pareto front of arrangement C-DSI-C.

almost pure product. Physical properties for the case of study are displayed in Table 2. We analyze four basic configurations obtained according to the rules of synthesis for hybrid systems proposed by Berry and Ng [7]: (i) hybrid distillation/melt crystallization processes using conventional distillation arrangement (Fig. 2); (ii) hybrid distillation/melt crystallization processes using an indirect thermally coupled distillation sequence (Fig. 3); (iii) hybrid distillation/melt crystallization processes using a modified arrangement from the indirect thermally coupled distillation sequence (distillation system studied by Tamayo-Galván et al. [15], Fig. 4); (iv) hybrid distillation/melt crystallization processes using a Petlyuk column (Fig. 5). For this class of systems, thermodynamic models such as NRTL can be used to calculate equilibrium vapour–liquid data and with temperatures and compositions of eutectic points, the activity parameters are calculated using the Wilson model in order to calculate the liquid–solid equilibrium [10]. The design pressure for each distillation arrangement was chosen to ensure the use of cooling water in the condensers. For the crystallizers, we assume that the separation of a pure isomer from the remaining melt at eutectic composition can be accomplished in one crystallization stage. In each crystallization region, one pure isomer crystallizes as product when the temperature is lowered in the crystallizer (those assumptions were also used in the work of Franke et al. [10]). In particular, for the simulation of the crystallizers in Aspen ONE Aspen Plus we use solid layer crystallization model. The parameters of this model were calculated based on operating conditions and solubility data of the mixture of isomers. The cooling energy is assumed to be equal to energy of crystallization and the volume of the equipment is calculated indirectly as a function of the amount of mixture being crystallized in the apparatus.

One important point to highlight is the selection of degrees of freedom for the equipment which were selected in function of the variables directly associated to the objective function. In this case the objective function selected was the minimization of the total annual cost. For example in the distillation column we choose as degrees of freedom reboiler duty and the number of stages because they are directly associated with the total annual cost.

6. Results

In this section, we first analyze the resulting Pareto fronts of the four hybrid configurations studied. Then we analyze the control properties of the studied schemes bases in singular value decomposition technique.

6.1. Hybrid configuration C-DSI-C

We calculate the Pareto front using the multiobjective genetic algorithm as design tool. Fig. 6 shows the Pareto front for case C-DSI-C, which includes the objectives to minimize, total heat duty and total number of stages of the distillation sequence, along with the total annual cost of the system (calculated with the information of the optimal design). It is important to remark that optimal designs satisfy the specified purities and recoveries in the distillation column and crystallizers; Figs. 7 and 8 show the profiles of

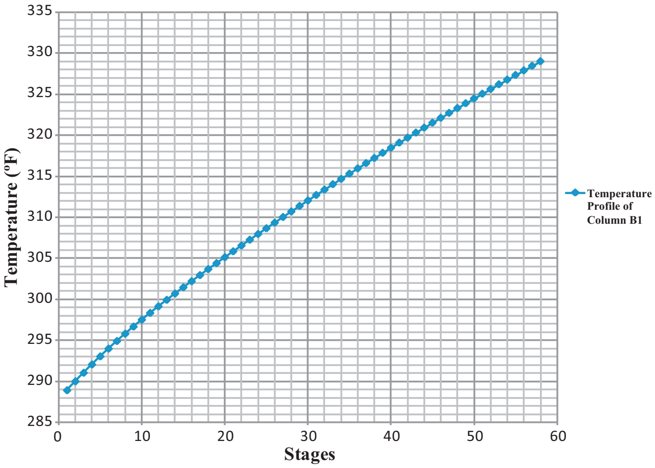


Fig. 7. Temperature profiles for the distillation columns in arrangement C-DSI-C with the minor total annual cost for column B1.

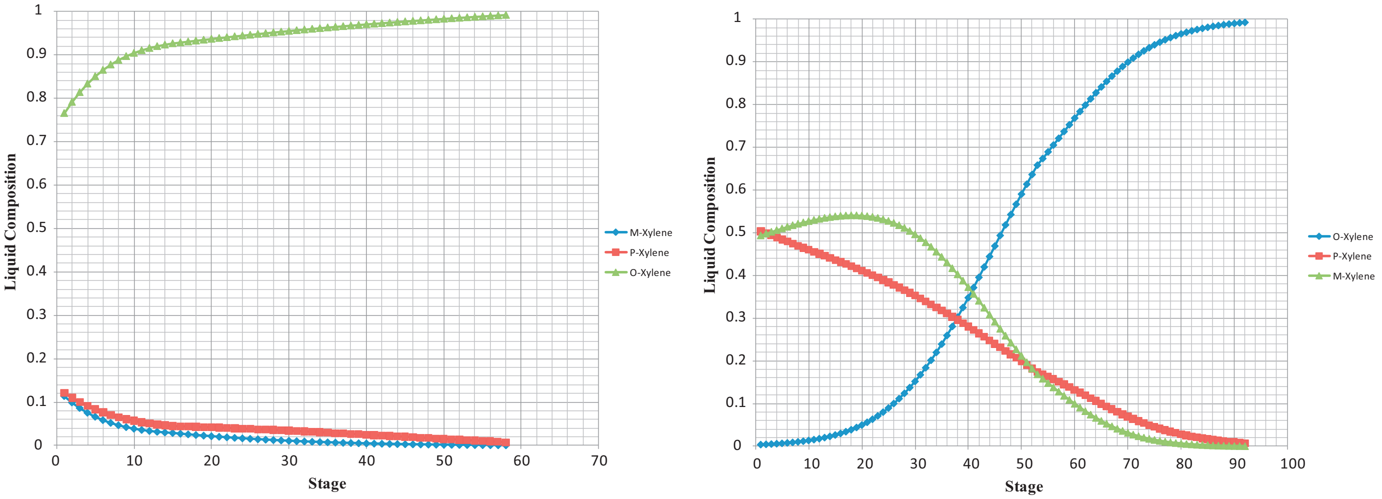


Fig. 8. Composition profiles for the distillation columns in arrangement C-DSI-C with the minor total annual cost: (a) column B1; (b) column B2.

temperature and composition for the distillation columns, respectively. The composition profiles in the O-xylene isomer meet the constraint imposed by genetic algorithms, obtained with a purity of 99% for the bottom of the first column (B1). In Fig. 8, it can be observed that in the top of the column B1 there is a ternary mixture of M-P xylene isomers with the rest of O-xylene that could not be separated in the first column. The second column (B2) gives the rest of O-xylene in a way that increases to 99% the purity at the bottom, and it gets a binary mixture of M-P xylene isomers in the top of the column with purities of 49.32% and 50.32% respectively in the liquid phase, complying with the optimization constraints. For further purification, obtained mixture is treated on the crystallizer. M and P xylene isomers have been obtained with purities of 99% in the crystallizers. A second observation is that the Pareto front shows a set of feasible designs, from where we highlight two: a design with minimum number of stages in the distillation columns and a design with minimum total annual cost, calculated using the method of Guthrie [38], and minimum reboiler duty in the distillation arrangement. The two possible designs contrast the objectives of minimizing the number of stages in the distillation columns (1,724,715 US$/yr) with minimizing the total annual cost and energy requirements in reboilers (1,410,871 US$/yr). Thus, the engineer can choose the best design for his particular needs. Each design in the Pareto front is an optimal design with different trade-offs between the objectives considered; and this set includes designs from minimum number of stages to minimum total annual cost, along with all designs between these extremes. Also, from Fig. 6 we can observe a good diversity in the designs that made up the Pareto front: number of stages and heat duties in the distillation system and total annual cost of the system covers a wide range of values. From Fig. 6 we also observed that energy requirements can be reduced in 17% approximately, just adding 20 stages to the sequence; this is an important observation since the choosing of the design must obey to economics of the process. Thereby, adding 20 stages is better if a long term saving in energy requirements is reached.

Tables 3 and 4 display parameters for the distillation columns and crystallizers for both optimal cases selected. The column tagged as “minimum on other variables” corresponds to a design with an optimal on energy requirements, which also represents a design with low TAC [39], high thermodynamic efficiency [26], low green house gas emissions [40] and, as a consequence, low environmental impact. Calculations for emissions of green house gases (particularly CO_2) are performed according to the method proposed by Gadalla et al. [40] Thermodynamic efficiency has been computed following the method reported by Gómez-Castro et al. [26]. As seen in Table 3, design with the lowest total annual cost required 20 stages more than the design with minimum number of stages in the distillation columns. On the other hand, the design with minimum number of stages in the distillation configurations used 27.87% more energy and showed an increment of 18.19% in the total annual cost that the most economical design.

6.2. Hybrid configuration C-TCDS-C

Fig. 9 presents the Pareto front of the arrangement C-TCDS-C. This figure highlights three designs that minimize a design objective. One is the design with fewer stages in the distillation columns, other design that minimizes the total annual cost of the system, and the last one represents the design that minimizes the energy requirements in the reboilers of the distillation columns. The design with minimum number of stages presents a total annual cost of 1,341,946 US$/yr; the configuration with minimum energy requirements in the reboilers shows a total annual cost of 1,212,290 US$/yr and the arrangement with minimum total annual cost presents a value of 1,206,375 US$/yr. It is also possible to observe that the design with minimum number of stages used 11 stages less than the design with minimum total annual cost and 21 stages less than the design with minimum energy requirements in the reboilers. On the other hand, the first design consumes 23.40% and 16.03% more energy than the second and third design respectively, and the total annual cost is 11.23% and 10.7% higher than those of the second and third design respectively. As for the application of process synthesis, although the first configuration minimizes the number of stages used in the separation is not a good design choice, since its cost and energy requirements are higher compared to those of the other two designs. The second design uses more energy but has a lower total annual cost in contrast to the third design, so for this particular case, the benefit in the selection of a separation scheme should be focused on minimizing the total annual cost. Therefore the design selected for comparative purposes, is the one with the lowest total annual cost. Composition profiles in hybrid design with a thermally coupled distillation column show that O-xylene isomer complies with the purity that serves as a restriction on genetic algorithms, obtaining 99% in the bottom of the first column (B1). The second column (B2) gives the rest of O-xylene, with a composition of 99% in the bottom, and a binary mixture of M-P xylene isomers in the top with purities of approximately 49.32%, 50.32% in the liquid phase composition respectively, which meet the constraints of optimization. It can be seen that the purities imposed in the crystallizers were

Table 3
Optimal design of distillation columns for arrangement C-DSI-C with lowest total annual cost and lowest number of stages in the distillation columns.

Distillation Sequences [DSI]	Design with the minimum on *N*	Design with the minimum on other variables
Number of Stages Column B1	43	58
Number of Stages Column B2	87	92
Reflux Ratio Column B1	22.03	16.39
Reflux Ratio Column B2	105.21	78.84
Feed Stage Column B1	16	15
Feed Stage Column B2	49	53
Heat Duty Column B1 (kJ/h)	8330593.57	5766829.33
Heat Duty Column B2 (kJ/h)	7660112.03	5766829.33
Pressure Design Column B1 (bar)	1.01	1.01
Pressure Design Column B2 (bar)	1.01	1.01
Bottom Pressure Column B1 (bar)	1.70	1.70
Bottom Pressure Column B2 (bar)	1.70	1.70
Temperature One Stage before the Condenser of Column B1 (K)	416.60	416.50
Condenser Temperature Column B1 (K)	415.80	415.90
Temperature One Stage before the Reboiler of Column B1 (K)	437.70	437.80
Reboiler Temperature Column B1 (K)	438.10	438.20
Temperature One Stage before the Condenser of Column B2 (K)	412.20	412.2
Condenser Temperature Column B2 (K)	411.90	411.90
Temperature One Stage before the Reboiler of Column B2 (K)	437.90	438.00
Reboiler Temperature Column B2 (K)	438.20	438.20
Diameter of Column B1 (m)	1.45	1.31
Diameter of Column B2 (m)	1.45	1.25
FEED1 (kmol/h)	35.26	35.26
DIST2 (kmol/h)	1.99	1.99
BOTT1 (kmol/h)	26.63	26.63
BOTT2 (kmol/h)	6.64	6.64
FEED2 (kmol/h)	8.63	8.63
Total Annual Cost (US$/yr)	1724715.92	1410871.56

Table 4
Optimal design of crystallizers for arrangement C-DSI-C with lowest total annual cost and lowest number of stages in the distillation columns.

Crystallizers	Design with the minimum on *N*	Design with the minimum on other variables
Operation Temperature Crystallizer C1 (K)	268.25	268.25
Heat Duty Crystallizer C1 (kJ/h)	−118428.39	−118428.39
Operation Pressure Crystallizer C1 (bar)	1.00	1.00
Volume Crystallizer C1 (m^3)	10.59	10.59
Residence Time (h)	1.00	1.00
Crystals production Masic Flow Crystallizer C1 (kg/h)	6877.78	6877.78
Magma Density at the outlet from Crystallizer C1 (kg/m^3)	649.74	649.74
Operation Temperature Crystallizer C2 (K)	270.65	270.65
Heat Duty Crystallizer C2 (kJ/h)	−55937.76	−55937.76
Operation Pressure Crystallizer C2 (bar)	1.00	1.00
Volume Crystallizer C2 (m^3)	0.22	0.22
Residence Time (h)	1.00	1.00
Crystals production Masic Flow Crystallizer C2 (kg/h)	106.11	106.11
Magma Density at the outlet from Crystallizer C2 (kg/m^3)	484.29	484.29
Total Annual Cost of Crystallizers (US$/yr)	184.23	184.23

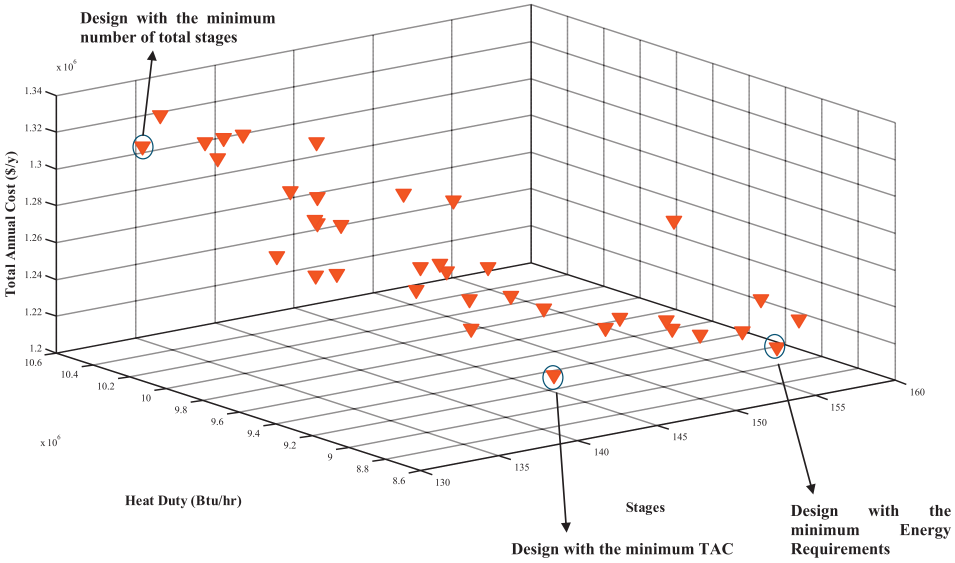


Fig. 9. Pareto front of arrangement C-TCDS-C.

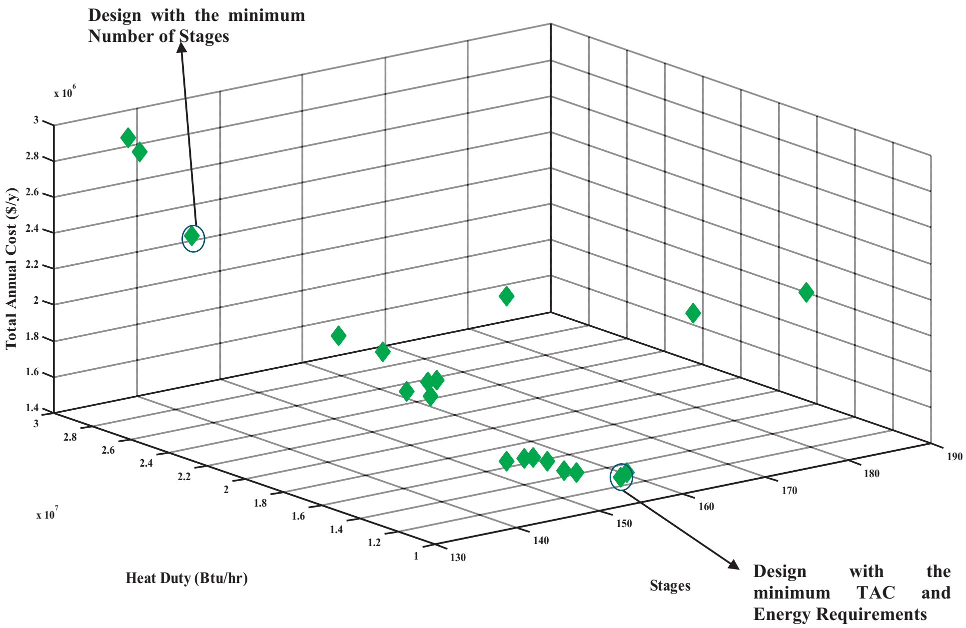


Fig. 10. Pareto front of arrangement C-MTCDS-C.

achieved. At this point it is possible to highlight that the best design of hybrid system with a thermally coupled distillation column shows a decrease of 14.5% in the total annual cost in comparison with the best design of the hybrid system with a system of conventional distillation columns.

6.3. Hybrid configuration C-MTCDS-C

The third hybrid system shows a distillation system, which is a modification of a thermally coupled distillation column. The presence of recycle streams in distillation columns with thermal coupling has influenced the notion of control problems during the operation of these systems with respect to the rather well-known behavior of conventional distillation sequences. For this reason, it has been proposed several alternate configurations to distillation schemes with thermal coupling, as the configuration discussed in this hybrid system, eliminating the recycle streams that appear to have some operational advantages over expected dynamic properties of the thermally coupled distillation sequences [16]. Fig. 10 shows the Pareto front with two designs that minimize an objective of the optimization, one is the design with minimum number of stages (2,038,549 US$/yr), and the other turns out to be the design that minimizes the total annual cost (1,405,896 US$/yr). In this case, the configuration with minimum number of stages shows that the total annual cost and energy requirements are 45.5% and 53% higher than those of the design with the lowest total annual cost. The design that has a minimum number of stages used 22 stages less than the design with the lowest total annual cost. For use in process

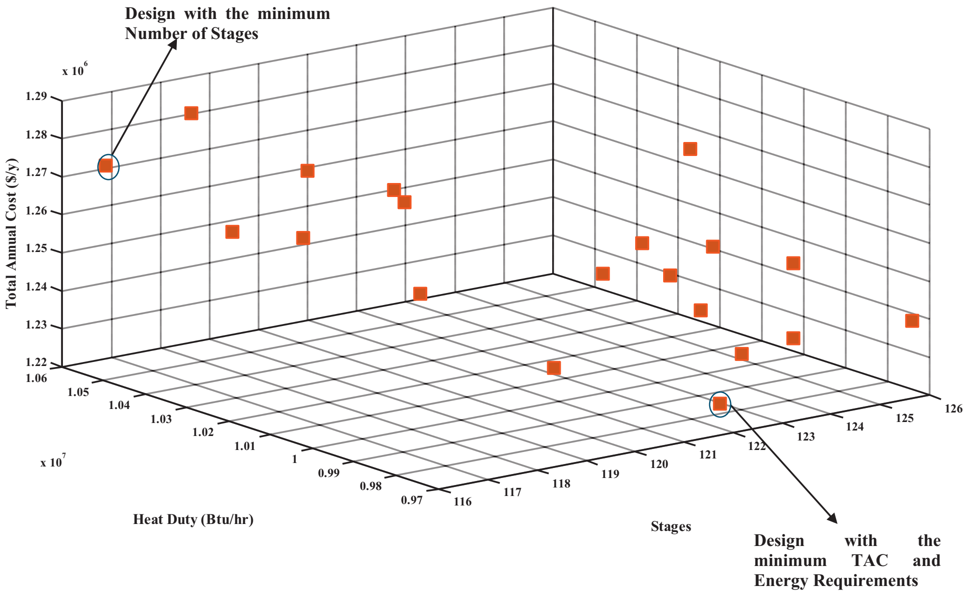


Fig. 11. Pareto front of arrangement C-PC-C.

Table 5
Transfer function matrix for system C-DSI-C.

$$G(s)=\begin{bmatrix} \dfrac{2.672}{1+0.828823s} & 0 & 0 \\ \dfrac{-34.5372}{1+3.034553s} & \dfrac{3.114}{(3.114)^2s^2+2(3.114)s+1} & \dfrac{-90.4744e^{-s}}{(2.484993062)^2s^2+2(2.484993062)s+1} \\ \dfrac{40.9324e^{-4.5s}}{(9.00300080)^2s^2+2(9.00300080)s+1} & \dfrac{-148.3976e^{-2s}}{(0.99769686)^2s^2+2(0.99769686)s+1} & \dfrac{-27.3884}{1+3.25831121s} \end{bmatrix}$$

synthesis, although the first configuration minimizes the number of stages used in the separation, not showing to be a good choice in economic terms and therefore discarded for further analysis. Also in this hybrid system purities as restrictions imposed on the genetic algorithm (99%) for the recoveries of the three isomers of xylene are achieved in the distillation system and crystallizers. When comparing the best design selected for the three hybrid systems that have so far been analyzed, it can be noticed that the best option is the

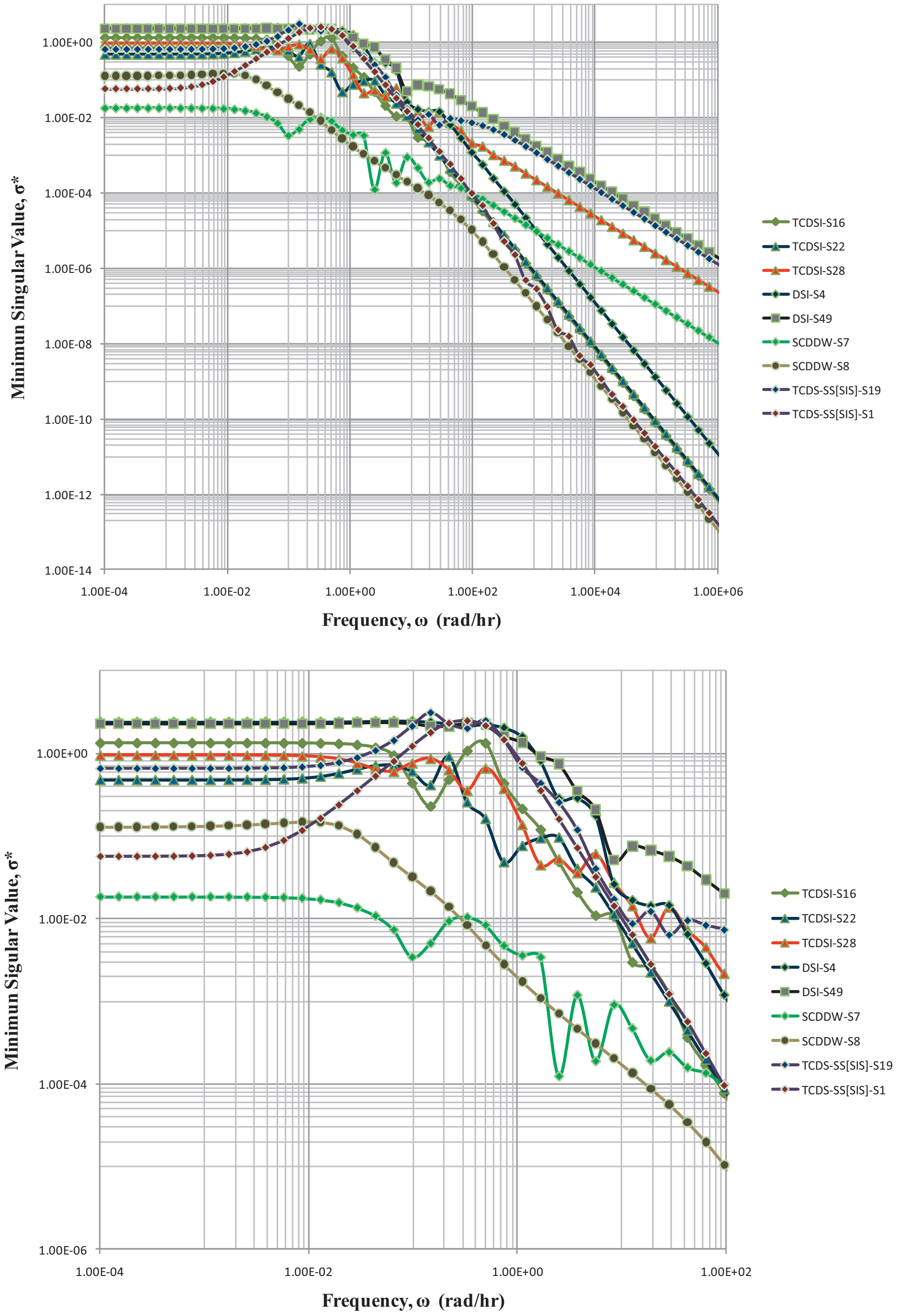


Fig. 12. Morari Resiliency Index values for optimal hybrid systems.

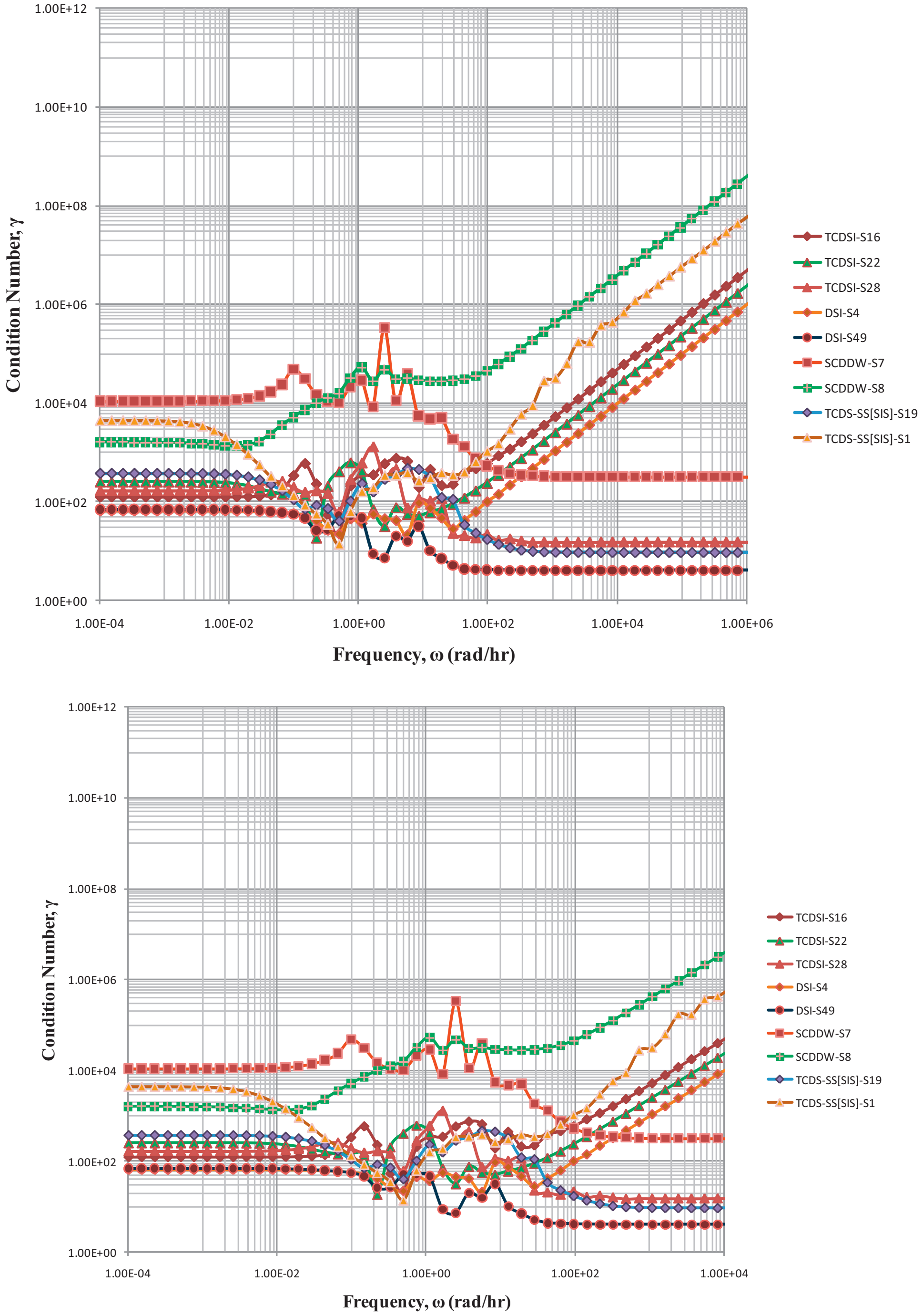


Fig. 13. Condition number for optimal hybrid systems.

setting that uses a thermally coupled distillation system, since it is the one showing the lowest total annual cost.

6.4. Hybrid configuration C-PC-C

Fig. 11 displays the Pareto front for the hybrid system with a Petlyuk distillation column. The Pareto front highlights two designs that minimize an objective, one of them is the design with minimum number of stages, and the other one is the design that minimizes the energy requirements in the reboiler of the Petlyuk column and the total annual cost of the system. The selected designs comply with the restrictions of the purities in the distillation column and the crystallizer. The first design has 6 stages less than the second design, but consumes 7.3% more energy and its total annual cost is 4% higher than the second. So the design selected for comparison is one that shows the lowest total annual cost and minimum energy requirements. The total annual cost of the design that turns out to be the cheapest is 1,226,443 US$/yr. At this point emerges an important point to highlight, the best hybrid design with a thermally coupled distillation column (C-TCDS-C) has a total annual cost of 1,206,375 US$/yr and a total number of stages in the distillation column of 170 while the best hybrid design with a Petlyuk column (C-PC-C) shows a total annual cost of 1,226,443 US$/yr with a total number of stages 132. The difference between the total annual cost between the system more affordable compared to the more expensive is 1.6%. This result implies that for practical purposes the energy requirements between the two systems is almost similar. However, the difference between the numbers of stages of the hybrid configuration with the lowest value compared with that shows the highest number is 28.8%. Given these results, it is possible to conclude that the best design among the four case studies reviewed is the hybrid system that includes a Petlyuk distillation column.

6.5. Controllability results

In order to complement the work, it was carried out a study of the dynamic properties of the design of optimal hybrid systems. We conduct a SVD analysis where is required open-loop transfer functions. In this study, step changes in the input variables were implemented and the open-loop dynamic responses were registered in Aspen Dynamics. An important point to note is that the Aspen Dynamic solve for the distillation columns equations presented as (8)–(12). That is the rigorous dynamic model for distillation columns. The dynamic responses were adjusted to transfer functions and arranged into transfer function matrices. Table 5 presents an example of a typical transfer function matrix for the case of separation analyzed. It can be noted that the dynamic responses can be adjusted to first or second order transfer functions with or without dead times.

Figs. 12 and 13 present the Morari resiliency index and condition number for optimal hybrid systems in the frequency domain. A clear trend in the parameters is observed: for this separation task, the hybrid system using a conventional distillation sequence shows the lowest value of γ and the highest value of σ^*. These results indicate that the hybrid system C-DSI-C has better theoretical control properties than the other hybrid systems using complex distillation sequences. However, the system C-TCDS-C shows similar values of γ and σ^* to the system C-DSI-C. It can be expected that the hybrid system using a thermally coupled distillation option will present similar closed-loop dynamic behavior for both set point tracking and load rejection to the hybrid system using a conventional distillation arrangement. In the case of the hybrid system using a Petlyuk distillation column, it can be seen that shows the worst control properties because it has the highest values of condition number and the lowest values of the Morari resiliency index (Figs. 12 and 13).

At this point it is important to note that according to the results obtained in steady state it can be concluded that the best design option is the hybrid system that uses a thermally coupled distillation column as it shows the lowest total annual cost and control properties similar to the system using a conventional distillation column (which is the one that shows the best control properties). Although the thermally coupled column has a larger number of stages in comparison with the hybrid system that uses a Petlyuk column, the total annual costs are very similar, but the hybrid design with a Petlyuk distillation column shows the worst control properties compared with the system C-TCDS-C. Thus making a comparative analysis the best design hybrid option is C-TCDS-C.

7. Conclusions

A strategy for the design and optimization of hybrid separation processes using genetic algorithms has been presented. As case of study, purification of a ternary mixture of close-boiling ortho, meta and para isomers of xylene is considered. The stochastic procedure allows manipulation of continuous and integer variables simultaneously. All resulting optimal designs considered full use of the equations of the models contained in the Aspen Plus simulator. The Pareto fronts obtained for hybrid systems present good diversity, in terms of the different structures of the columns, and also with respect to energy requirements. Moreover, it has been found that the optimum energy requirements design can be related to the minimum total annual operating cost. According to the results obtained in steady state is concluded that the best design option is the hybrid system that uses a thermally coupled distillation column (C-TCDS-C) as it shows the lowest total annual cost and control properties similar to the system using a conventional distillation column (which is the one that shows the best control properties). The benefit of our approach lies in the ability to deal with complex hybrid flowsheets and design of the optimal process with robustness and efficiency.

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