



The importance of the sequential synthesis methodology in the optimal distillation sequences design

Massimiliano Errico^{a,*}, Ben-Guang Rong^b, Carlo Edgar Torres-Ortega^c,
Juan Gabriel Segovia-Hernandez^c

^a Università degli Studi di Cagliari, Dipartimento di Ingegneria Meccanica, Chimica e dei Materiali, Via Marengo 2, 09123 Cagliari, Italy

^b University of Southern Denmark, Department of Chemical Engineering, Biotechnology and Environmental Technology, Niels Bohrs Allé 1, DK-5230 Odense M, Denmark

^c Universidad de Guanajuato, Campus Guanajuato, Division de Ciencias Naturales y Exactas, Departamento de Ingeniería Química, Noria Alta S/N, Guanajuato 36050, Mexico

ARTICLE INFO

Article history:

Received 4 July 2013

Received in revised form

12 November 2013

Accepted 14 November 2013

Available online 2 December 2013

Keywords:

Distillation

Design

Optimization

Simulation

Process synthesis

Process modeling

ABSTRACT

The sequential design method is presented as a complementary tool of the systematical synthesis procedure that allows to define a clear connection among the different types of distillation column sequences. In particular, the connection with the simple column subspace is considered, since this subspace represents the comparison reference for all the alternatives considered. The sequential design procedure, based on the correspondence between the functionality of the column's section among the simple columns and the derived sequences, is compared with a mathematical based optimization algorithm. The separations of a four-component near ideal mixture and the azeotropic ethanol–water mixture are considered as case studies and the designs obtained applying both methods have been compared. The results confirmed that the sequential design method is a fast and reliable tool in the optimal design of the column sequence.

© 2013 Elsevier Ltd. All rights reserved.

1. Introduction

The definition of a searching space that includes all or most of the possible alternatives for the separation of a multicomponent mixture, is the first step in the identification of the optimal flow-sheet. Once the most suitable separation method is identified, the problem becomes how to generate the alternatives to be included in the searching space. The most suitable separation method is usually selected by using appropriate indexes. When the relative volatility between the key components is higher than 1.05, distillation can be considered as a feasible separation method (Seader & Westerberg, 1977). Focusing on distillation, the well known simple column sequences are the first class of configurations explored. They represent the simplest solution since are composed by columns with a single feed, one top and one bottom product. Anyway their design simplicity is counterbalanced by the high energy consumption and high capital costs.

The high energy consumption is related to thermodynamic inefficiencies like the irreversible mixing of non-identical streams (feed mismatching) or the energy lost when a middle component is initially separated and then mixed again with the other components (remixing effect). The capital costs are mainly associated to the number of columns and auxiliary equipments. For a N -component mixture, if N product streams are required, it is necessary a sequence composed by $N - 1$ columns.

The number of simple column sequences can be predicted using the formula (Thompson & King, 1972):

$$S_N = \frac{[2(N - 1)]!}{[N!(N - 1)!]} \quad (1)$$

Among all the simple column sequences the most promising one/s, identified by means of predefined criteria, is/are utilized as reference case to compare the performances of all the other alternatives included in the searching space. A complete searching space could theoretically include sequences with more than $N - 1$, exactly $N - 1$ and less than $N - 1$ columns.

The convenience to include in the searching space configurations with less than $N - 1$ columns has been explored by Kim and Wankat (2004) for different four-component mixtures. For each feed composition case, the corresponding five simple column

* Corresponding author. Tel.: +39 070 6755079; fax: +39 070 6755067.

E-mail address: massimiliano.errico@dimcm.unica.it (M. Errico).

Nomenclature

Az.	azeotropic mixture
b_i	molar availability function of the output stream i [kJ mol ⁻¹]
b_l	molar availability function of the input stream l [kJ mol ⁻¹]
C	column
G_{\max}	maximum number of generations
h	molar enthalpy [kJ mol ⁻¹]
LW	lost work [kJ h ⁻¹]
N	number of feed components
n_i	molar flowrate of the output stream i [mol/h]
n_l	molar flowrate of the input stream l [mol/h]
PS	population size
Q_j	heat flow j out the system [kJ h ⁻¹]
Q_m	heat flow m in the system [kJ h ⁻¹]
s	molar entropy [kJ K ⁻¹ mol ⁻¹]
S_N	number of possible simple column sequences
TAC	total annual cost [k\$ yr ⁻¹]
T_0	temperature of surroundings [K]
$T_{s,j}$	temperature of the reservoir for the heat flow j out the system [K]
$T_{s,m}$	temperature of the reservoir for the heat flow m in the system [K]
vap.	vapor phase
W_{\min}	minimum work of separation [kJ h ⁻¹]
W_{\min}	shaft work k produced by the system [kJ h ⁻¹]
$W_{s,k}$	shaft work n fed to the system [kJ h ⁻¹]
ΔS_{irr}	irreversible entropy production [kJ K ⁻¹ h ⁻¹]
η	thermodynamic efficiency

sequences were considered and compared to the eleven alternative sequences proposed. Among all the alternatives, ten are composed by two columns and one alternative includes only one column with two side streams. Among the ten column sequences, two of them have a product side stream connecting the columns. All the alternative sequences were defined considering the possibility to have a side stream above or below the column's feed, in liquid or vapor phase. The authors considered the synthesis and the design as sub sequential steps. It means that once all the alternatives were identified, their design was done independently since no connection between the simple column sequences and the configurations included in the searching space was evidenced.

Caballero and Grossmann (2004) introduced a superstructure optimization approach able to generate a very complete set of distillation sequences that constitutes the searching space used by the authors to identify the configuration with the lowest cost. Simple column sequences, fully thermally coupled sequences, all the possible partially thermally coupled sequences and even the thermodynamic equivalent sequences were considered. An appropriate design method was selected to define the design parameters used in the cost evaluation. Once a feed composition case has been defined, the design calculation was performed for all the feasible sequences generated.

Shenvi, Shah, Zeller, and Agrawal (2012) completed the previous work of Shah and Agrawal (2010) introducing a eight-step matrix-based method able to include in the searching space also the configurations with less than $N-1$ columns. The matrix-based method proposed, includes both sharp and non-sharp splits and generates simple and thermally coupled configurations. The authors proved the validity of their methodology by means of a case study of a four-component normal paraffin mixture. All the configurations were first screened using a short-cut method, and

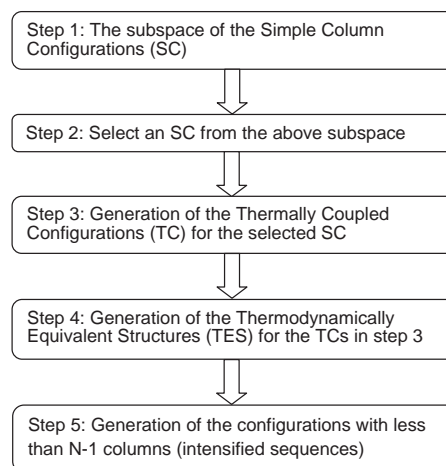


Fig. 1. The simplified systematic procedure for the sequential distillation sequence synthesis.

then the most promising ones were simulated again through a rigorous method. Since each class of configurations was developed individually, the design was done independently.

It is clear that all the authors approached the necessity to generate a complete searching space following different mathematical tools; anyway the simple column sequences are always considered as a basis for the comparison with the alternative sequences proposed.

The aim of this work is to analyze the benefits in the design procedure when the searching space generation method is able to keep a strict connection between the different sequences. Two case studies have been presented to prove the validity of the methodology. The first case regards the separation of a four-component hydrocarbon mixture; the second one is the well known ethanol–water azeotropic separation. For the same cases a hybrid multi-objective optimization algorithm is used to obtain the optimal design parameters. The results are then compared and commented.

2. The sequential design method

In our previous works (Errico & Rong, 2012a; Errico, Rong, Tola, & Turunen, 2009; Rong & Errico, 2012) the synthesis of alternative configurations was realized following a sequential method that allows to establish a clear connection between all the sequences. Fig. 1 reports a simplified five-step procedure that guides the designers from the simple to the complex configurations. The introduction of one or more thermal couplings is used to move from step 2 to step 3, then by the column section recombination the step 4 can be completed. The configurations included in the last step 5 are obtained by replacing one or more single column sections with a product side stream or an interconnecting column stream (intensified sequences).

Considering as an example the separation of a four component mixture, where the components are listed in decreasing order of relative volatility, the indirect-direct configuration reported in Fig. 2(a) was chosen among all the simple column configurations. The corresponding thermally coupled configurations can be obtained by replacing a condenser, a reboiler, or both, associated to non-product streams, with two interconnecting vapor and liquid streams between the columns. In the case considered three alternatives are possible, but only the one obtained by substitution of the condenser corresponding to the ABC submixture is reported in Fig. 2(b). The column sections, defined according to Hohmann, Sander, and Dunford (1982), are enumerated in each sequence. The thermodynamically equivalent configuration can be obtained for

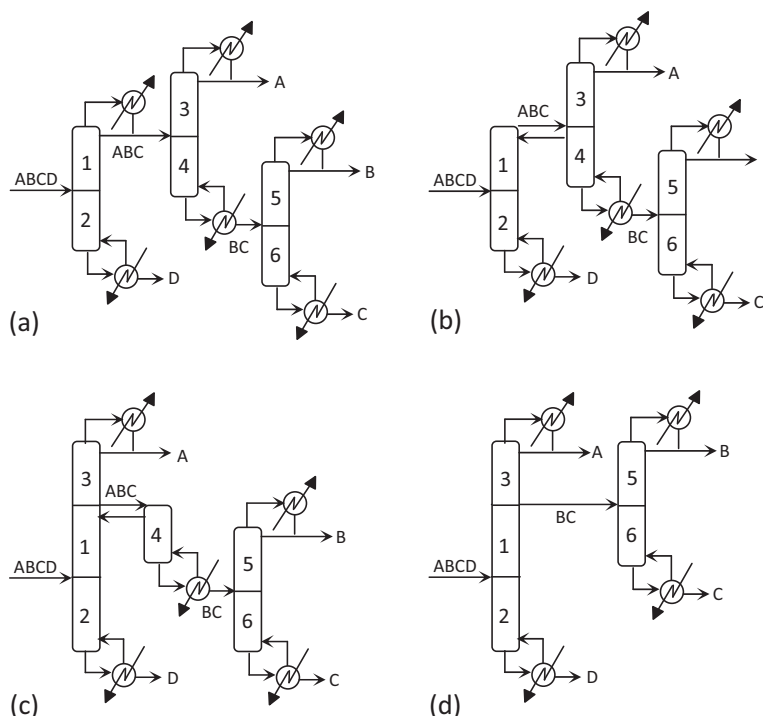


Fig. 2. Distillation sequences alternatives for a four-component mixture: (a) indirect-direct simple column sequence, (b) thermally coupled sequence, (c) thermodynamic equivalent sequence, (d) intensified/MSC sequence.

the corresponding thermally coupled sequence by recombination of the section associated to the thermal coupling. Fig. 2(c) was obtained moving section 3 above section 1. Finally Fig. 2(d) reports the case of the intensified sequence obtained by elimination of the transport section 4. It is clear that the configurations in Fig. 2(a) and (d) are related, since the column sections in both the configurations performed the same separation task. This approach has the benefit to generate a complete searching space and at the same time each alternative sequence can be easily associated to the “root” simple column sequence.

The sequential design method (SDM) can be defined as a column sequence design methodology deeply related to the synthesis method utilized to predict the configurations included in the searching space. This novel approach represents a clear benefit in terms of sequence design. As discussed in the introduction, the subspace including all the simple column sequences is always designed (Giridhar & Agrawal, 2010) and the best sequence selected is kept as reference for the alternative configurations. Thus, in all the cases, the design parameters for the simple column sequences are known. When the designer is interested to a configuration like the one reported in Fig. 2(d); its design is speeded up by the correspondence of the section’s parameters with the indirect-direct configuration already simulated. A final tuning allows the designer to obtain the optimal design in a reasonable time. Even if it is difficult to define the time saved, because mainly depends on the ability of the designer with the simulation tool used, it is possible to surely assert that is faster to obtain the final design when near-optimal parameters are used in the design initialization.

3. The multi-objective optimization algorithm

If the correspondence between the simple column sequences and the alternatives generated is unknown the design is done independently for each configuration using, for example, a pure mathematical approach.

Chemical engineering problems generally involve continuous variables with or without integer variables, usually formulated as a mixed integer non-linear programming problem (MINLP). All these characteristics make challenging the research of the global optimum. Stochastic global optimization (SGO) techniques are well-suited for such problems. Deterministic methods utilize analytical properties (e.g. convexity) of the optimization problem to generate a deterministic sequence of points (i.e. trial solutions) in the searching space, that converges to a global optimum. However, they require some assumption for their success and provide convergence guarantee only for the problems satisfying the underlying assumptions.

SGO techniques involve probabilistic elements and consequently use random numbers in the search for the global optimum. Thus, the sequence of points depends on the seed used for random number generation. In theory, SGO techniques need infinite iterations to guarantee convergence to the global optimum. However, in practice, they often converge quickly to an acceptable global optimal solution. In general, SGO techniques offer several attractive features:

- 1) They are simpler to understand and programming compare to deterministic methods.
- 2) They require no assumption in the optimization problem (e.g. continuity of the objective function and constraints), and hence can be used for any type of problem.
- 3) SGO methods are robust for highly nonlinear problems even with large number of variables
- 4) They often converge to (near) global optimal solution quickly
- 5) They can be adapted for non-conventional optimization problems. For example, several SGO techniques have been modified for multi-objective optimization (Rangaiah, 2010).

They appear to be a suitable alternative for the design and optimization of separation schemes (Martínez-Iranzo, Herrero, Sanchis, Blasco, & García-Nieto, 2009).

Several heuristic techniques for global optimization mimicking biological evolution have emerged in the literature. Among the most representative algorithms, the genetic algorithms (GA) (Golberg, 1989), the particle swarm optimization (PSO) (Kennedy & Eberhart, 1995), and a new class of evolutionary methods called differential evolution (DE) algorithms (Storn & Price, 1997) can be mentioned. For different theoretical and practical problems, comparative studies have shown that the performance of DE algorithms is clearly better than that of the other GA and PSO algorithms (Ali & Torn, 2004; Panduco & Brizuela, 2009; Sacco, Henderson, Rios-Coelho, Ali, & Pereira, 2009) in cases like the modeling of power systems, scheduling, the dynamic economic dispatch, electromagnetism and antenna problems, nuclear reactor design, classic benchmark problems, among others.

The optimization procedure chosen is based on the work of Sharma and Rangaiah (2010) where a Hybrid Multi-Objective (HMO) algorithm is used to screen all the possible design alternatives. The principles of this method are here briefly recalled. The HMO method is based on differential evolution with taboo list (MODE-TL). Specifically, MODE-TL includes classical differential evolution steps, adaptation for multiple objectives (selection of individuals for subsequent generations), taboo list and taboo check, and a convergence criterion based on the number of generations. According to Sharma and Rangaiah (2010), a population of NP individuals with D-dimension (number of decision variables) is initialized randomly inside the bounds on decision variables. A mutant vector is generated by adding the scaled difference of two randomly selected individuals with another randomly chosen individual. The elements of this mutant vector compete with those of the target vector, with probability Cr to generate a trial vector. Taboo list concept of Taboo Search is used in the multi-objective optimization method to avoid the revisit of the searching space by keeping the record of recently visited points. Taboo list is randomly initialized using the initial population and continuously updated with the newly generated trial individuals. This taboo check is implemented in the generation step of the trial vector, and the new trial individual is generated repeatedly until it is not near to any individual in the taboo list. Objective functions and constraints are evaluated for this new trial individual. A total of NP trial individuals are generated by the repetition of steps described. The newly generated NP trial vectors are combined with the parent population to form a combined population with 2NP total individuals. This combined population undergoes non-dominated sorting and ranking accordingly. Individuals with the same non-dominated rank are further ranked on the basis of crowding distance. The first (best) NP individuals are used as the population in the subsequent generation. The optimal design of the analyzed configurations was found by using HMO algorithm as an optimization method implemented in Microsoft Excel and coupled with Aspen Plus simulations. During the evolution of the HMO algorithm, the vector values of decision variables are sent from Microsoft Excel to Aspen Plus using dynamic data exchange by COM technology. These values are attributed in Microsoft Excel to the corresponding process variables and then sent to Aspen Plus. Note that using the COM technology it is possible to add a code such that the applications behave as an Object Linking and Embedding automation server. After running the rigorous simulation, Aspen Plus returns to Microsoft Excel the vector of results. Finally, Microsoft Excel analyzes the objective function value for the HMO procedure. In this case the total annual cost (TAC) and the thermodynamic efficiency (η) were used as objective functions. The TAC was calculated using the Guthrie's method as presented by Turton, Bailie, Whiting, and Shaiwitz (2004).

$$\text{TAC} = \sum \left[\left(\frac{\text{Capital Cost}}{\text{Time of Investment}} \right) + (\text{Cost of Utilities}) \right] \quad (2)$$

The column's shell, sieve trays, floating-tube & shell-condensers, Kettle-reboilers and multiple-pipes coolers were considered in the capital costs evaluation. A modular approach was used, that is the equipment base costs were estimated by the respective unit capacity, for instance, the shell by using the volume of the column, the heat exchangers by the heat transfer surface, and so on. The investment time was ten years. Finally steam and cooling water were considered as utilities.

The thermodynamic efficiency was computed using the thermodynamic laws for steady-state flow systems, Fig. 3 (Seider, Seader, Lewin, & Widagdo, 2009).

$$\sum_{in} \left[n_i b_i + Q_j \left(1 - \frac{T_0}{T_{s,j}} \right) + W_{s,k} \right] - \sum_{out} \left[n_l b_l + Q_m \left(1 - \frac{T_0}{T_{s,m}} \right) + W_{s,n} \right] = LW \quad (3)$$

$$b = (h - T_0 S) \quad (4)$$

$$LW = T_0 \cdot \Delta S_{irr} \quad (5)$$

$$\eta = \frac{-W_{min}}{(-LW - W_{min})} \quad (6)$$

All the thermodynamic properties, like enthalpies and entropies of the streams, were evaluated through the use of the process simulator Aspen PlusV7.3 for all the input and output streams.

4. Methods comparison

In order to compare the two different design approaches, two case studies already reported in the literature were considered. The first regards the separation of a near ideal four-component mixture, the second the separation of an ethanol–water stream. An ideal and a non-ideal system were selected to prove the generality of the design method. All the simulations were carried out using an Intel(R) Core(TM) i7 processor (3.40 GHz) with 12 GB RAM in Windows 7 Enterprise environment. The optimization by HMO was carried out focusing on the two objective functions (TAC and thermodynamic efficiency) already described. It is important to remark that the TAC includes the energy consumptions (around 75–90% of the total cost) and the capital costs (10–25%). Moreover, the thermodynamic efficiency was selected in order to study the effect of the thermal coupling, the internal mass recycles and the external solvent effects for different TACs, that means for different energy consumptions.

4.1. Case study 1

A particular subspace of distillation sequences was recently identified by Rong and Errico (2012). These sequences are characterized by similar features compared to the simple column configurations; each column is equipped with a condenser and a reboiler and the products are never obtained as side streams. It was evidenced that only some simple column configurations can generate these kinds of sequences named modified simple column (MSC) configurations.

Considering the case of four component mixtures the corresponding MSC configurations were considered by Errico and Rong (2012a). The energy consumption and the capital costs were compared to the corresponding best simple column sequence and to the simple column sequence from which are derived. In the present study the composition case reported in Table 1 is reconsidered. A feed flowrate of 100 kmol/h as saturate liquid and at the pressure of 4.6 bar is fed to the first column. The product purities were set, on

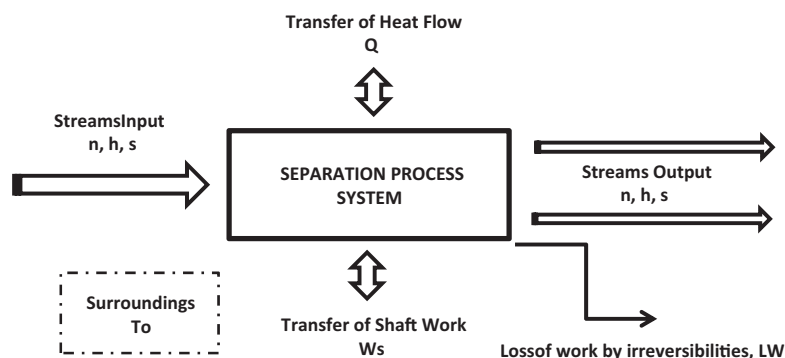


Fig. 3. Thermodynamic analysis of a separation process system.

Table 1

Feed composition for case study 1 and 2.

Component	Composition (%mol)	
	Case study 1	Case study 2
<i>n</i> -Butane	5	–
Benzene	10	–
<i>n</i> -Heptane	10	–
<i>n</i> -Nonane	75	–
Ethanol	–	5
Water	–	95

molar basis, to 99.2% for the *n*-butane, 97.0% for the benzene, 97.3% for the *n*-heptane and 99.7% for the *n*-nonane. The simulations were performed by means of Aspen Plus V7.3. BK10 was selected as thermodynamic property method and the rigorous stage by stage model RadFrac was used to model the distillation columns. Equilibrium stages were considered.

The simple column configuration of Fig. 2(a) and the corresponding MSC configuration of Fig. 2(d) are studied. The design of the MSC configuration was defined considering the correspondence between the sections function in the simple column and the MSC sequence. Table 2 describes the main specifications used for the HMO algorithm. The maximum number of generations (G_{max}) and the population size (PS), selected after a slightly tuning process was 70 and 80, respectively. The configuration parameters, the energy consumption and the capital costs evaluation for the reference and the MSC configurations designed with the sequential method and the optimization algorithm are reported in Table 3. Should be noted that since the capital cost evaluation in the HMO method was performed using the Guthrie's method, for the final design the calculations were repeated using the Economic Evaluator implemented in Aspen Plus. The HMO algorithm optimizes 7 parameters: the number of stages, the feed location, the reflux ratio for both

Table 2

Specifications of HMO for the case study 1.

Decision variable	Lower boundary	Upper boundary
Total trays C_1	20	70
ABCD tray	10% ^a	95% ^a
BC output tray	10% ^b	95% ^b
Total trays C_2	20	90
BC input tray	10% ^c	95% ^c
Reflux ratio C_1	15	30
Reflux ratio C_2	1	10

^a Position of the stream tray expressed as percentage of the total number of column's trays. The ABCD stream can vary from the first to the last column tray.

^b Position of the stream tray expressed as percentage of the total number of column's trays. The BC stream is located above the feed of the first column.

^c Position of the stream tray expressed as percentage of the total number of column's trays. The feed location of the second column can vary from the first to the last column tray.

columns, and the side stream location. Only the configurations that satisfy the purity constraints are considered for the economic evaluation. The final configuration was obtained after 15 h and globally 5600 simulations were examined. Among all the solution examined 4733 individuals satisfied the purity targets imposed by the problem.

The Pareto front diagram reported in Fig. 4 shows a clear competition between the objective functions that makes the optimization harder. This fact can be explained considering that decreasing the energy consumption, the thermodynamic efficiency increases until some level, where after that, the capital costs starts growing significantly with just small reductions in the energy consumption.

The corresponding capital cost and energy consumption were reported in Table 3. The filled circle point in Fig. 4 represents the selected configuration from the HMO algorithm and used to compare the results with the SDM.

From the results reported in Table 3 it is possible to notice that the results obtained with both design methods are very similar, only a slight increase of the heat loads and the capital costs are observed for the HMO design. In particular the total condenser and reboiler duties together with the capital costs are 5%, 4% and 6% higher than the values obtained applying the SDM. The performance of HMO could be explained on base that the initial values, and/or the number of generations and/or the population size were not properly selected.

4.2. Case study 2

The second case study is about an azeotropic separation for the production of bioethanol. Considering the importance in developing cost effective flow sheets, many studies are focused on the generation of alternative schemes especially when distillation is considered as separation method (Aviles Martinez et al., 2012; Errico & Rong, 2012b; Taylor & Wankat, 2004). Recently, different alternatives for the bioethanol separation by extractive distillation are considered by Errico, Rong, Tola, and Spano (2013). As done for the ideal mixtures, also in this case, the new sequences proposed have been related to the traditional design composed by three simple columns and reported in Fig. 5(a).

In that configuration the first column, named pre-concentrator, is necessary to approach the azeotropic composition, the distillate and the solvent are fed to the extractive distillation column where ethanol at the desired purity is obtained. The last column of the sequence recovers the solvent from the bottom and a water-ethanol stream as distillate. This stream is recycled back to the pre-concentrator in order to increase the ethanol and water recovery. A flowrate of 1694.24 kmol/h as saturate liquid, at the pressure of 1 bar and with the composition reported in Table 1 is fed to the pre-concentrator. The product purity requirements are, on molar basis, 99.9% for the ethanol and 99.9% for the water stream,

Table 3
Comparison between the design methods for case study 1.

	Fig. 2(a)			Fig. 2(d)			
	C ₁	C ₂	C ₃	Sequential design		Optimization algorithm	
	C ₁	C ₂	C ₃	C ₁	C ₂	C ₁	C ₂
Number of stages	31	28	67	45	67	40	71
Feed stage	15	14	35	29	35	27	41
Side stream stage	–	–	–	14	–	11	–
Reflux ratio	1.94	1.37	3.79	21.64	3.79	23.83	3.64
Column diameter (m)	0.94	0.26	0.53	0.94	0.53	0.94	0.51
Total condenser duty (kW)		–1059.30			–956.05		–1003.72
Total reboiler duty (kW)		1332.12			1228.78		1276.71
Annualized capital cost (k\$yr ⁻¹)		57.748			51.038		54.050
Thermodynamic efficiency (%)		32.00			32.60		32.67

Table 4
Comparison between the design methods for case study 2.

	Fig. 5(a)			Fig. 5(d)			
	C ₁	C ₂	C ₃	Sequential design		Optimization algorithm	
	C ₁	C ₂	C ₃	C ₁	C ₂	C ₁	C ₂
Number of stages	44	28	9	44	36	47	51
Reflux ratio	2.337	0.149	0.580	2.337	0.197	1.91	0.33
Distillate flowrate (kmol h ⁻¹)	103.375	85.037	18.338	103.375	85.011	103.946	85.000
Feed stage	30	25	3	30	25	31	28
Vapor recycle feed stage	34	–	–	34	–	40	–
Solvent feed stage	–	3	–	–	3	–	3
Total solvent feed	–	–	–	–	54.3	–	38.32
Column diameter (m)	1.37	0.74	0.65	1.37	0.79	1.28	0.84
Total condenser duty (kW)		–3820.645			–3750.638		–3422.695
Total reboiler duty (kW)		5018.671			4934.136		4392.951
Solvent makeup (kmol h ⁻¹)		0.001			0.010		0.060
Annualized capital cost (k\$yr ⁻¹)		134.3			112.9		118.4
Thermodynamic efficiency (%)		10.68			12.62		14.24

moreover it is necessary to minimize the fresh solvent flowrate maximizing its recovery in the second column. Glycerol was considered as solvent and the NRTL method was utilized to evaluate the activity coefficients. The rigorous model RadFrac, already implemented in Aspen Plus was considered together with equilibrium stages. The convergence of the system was improved selecting the sum-rates method in the convergence option data sheet.

Applying the thermal coupling principle it is possible to generate the thermally coupled sequence reported in Fig. 5(b), then recombining section 8, the thermodynamic equivalent configuration in Fig. 5(c) is obtained. The possibility to reduce the number of columns is reported in Fig. 5(d) where only two columns are employed to carry on the separation. Applying the SDM and HMO

algorithm the configurations are compared for the design parameters, the energy consumption and the capital costs.

The results are reported in Table 4. Since the SDM utilizes the parameters obtained from the root configuration of Fig. 5(a), these are also reported in the aforementioned Table. The HMO algorithm optimizes 11 parameters: the number of stages, the feed location and the reflux ratio for both columns, the vapor recycle stream to the first column, the solvent feed stage and the vapor side stream tray of the second column, the total solvent flowrate and the amount of solvent recovered in the second column's bottom stream, as an extra cost the glycerol make-up was considered. The final solution was obtained after 33 h, 7200 simulations were evaluated and 3737 individuals satisfy the problem's purity targets.

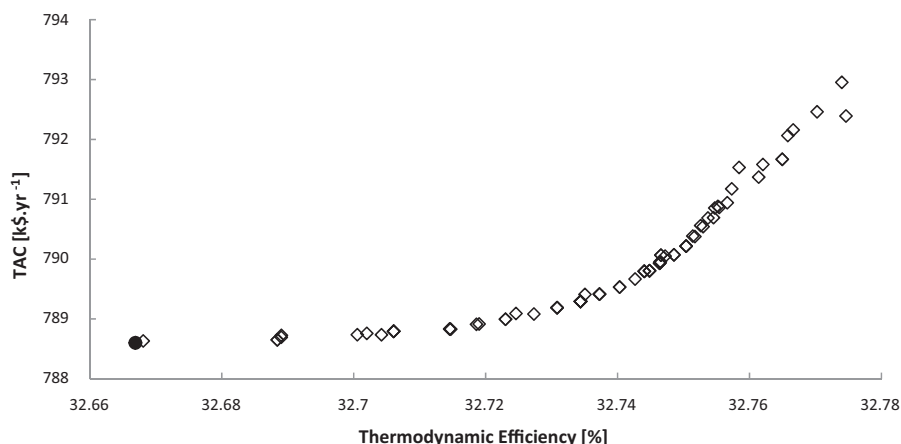


Fig. 4. Pareto front for the case study 1. The filled point corresponds to the solution compared with the SDM.

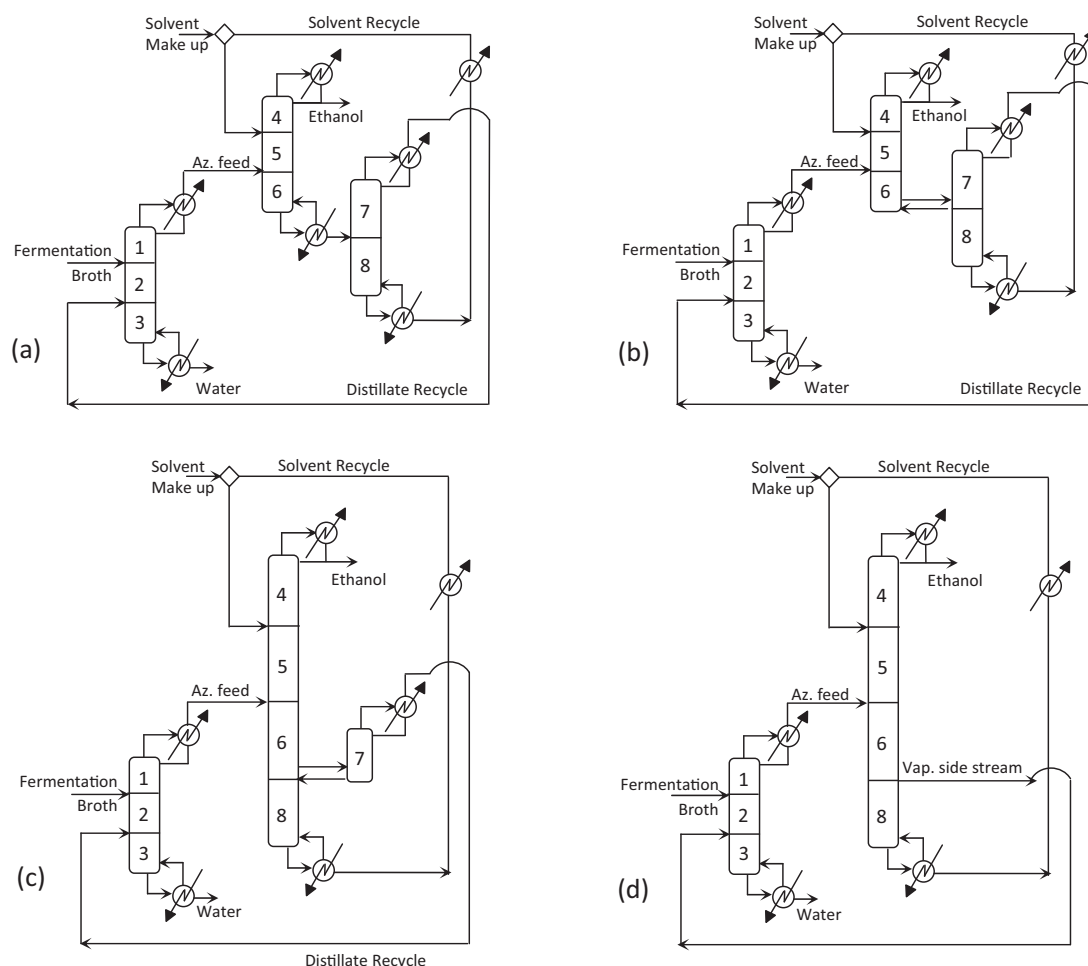


Fig. 5. Separation sequences by extractive distillation for ethanol production: (a) traditional sequence, (b) thermally coupled sequence, (c) thermodynamic equivalent sequence, (d) two-column sequence.

Table 5 describes the specifications used for the HMO algorithm. The G_{\max} and PS selected after a slightly tuning process were 60 and 120, respectively.

The corresponding Pareto front is reported in **Fig. 6**. The dot selected (depicted by the filled circle point) corresponds to the lowest TAC value and, in this case, to the highest thermodynamic efficiency.

Table 5
Specifications of HMO for the case study 2.

Decision variable	Lower boundary	Upper boundary
Total trays column I	15	60
Fermentation broth tray	30% ^a	69.60%
Vap. side stream (input) tray	40%	95%
Reflux ratio column I	0.5	6
Total trays column II	10	60
Az. feed tray	40% ^a	85% ^a
Vap. side stream (output) tray	20% ^b	80% ^b
Solvent tray	5% ^c	40% ^c
Vap. side stream flow (kmol/h)	19	25%
Solvent flow (kmol/h)	38	63%
Reflux ratio column II	0.05	2

^a Position of the stream tray expressed as percentage of the total number of column's trays. The feed stream can vary from the first to the last column tray.

^b Position of the stream tray expressed as percentage of the total number of column's trays. The vap. stream is located below the feed of the second column.

^c Position of the stream tray expressed as percentage of the total number of column's trays. The solvent is fed above the second column's feed.

The corresponding capital cost, and energy consumption were reported in **Table 4**. As already noticed for the previous case study, there is a competition between the objective functions even if the general trend appears to be more linear. Moreover, the TAC seems to decrease by increasing the thermodynamic efficiency. This opposite behavior with respect to the case study 1 reflects the complex effects of more recycles and the presence of an external solvent that allows higher energy consumptions, therefore higher thermodynamic efficiencies.

From a first analysis of the results reported in **Table 4** it is possible to notice that the design obtained with the HMO method slightly outperforms the results obtained with the SDM. The total condenser and reboiler duty are respectively 9% and 11% lower, contrarily, the capital costs for the configuration designed with the SMD are 5% lower than the correspondent value obtained with the HMO.

On the other hand the make-up flowrate necessary to replenish the solvent losses in the HMO design is six times the value obtained with the SDM. According with [Garcia-Herreros, Gomez, Gil, and Rodriguez \(2011\)](#) the glycerol's cost was estimated at 75 \$ kmol⁻¹, considering a mean operational time of 8000 hr⁻¹, the corresponding cost is 6.0k\$yr⁻¹ for the SD and 36.0k\$yr⁻¹ for the design obtained with the HMO. This difference can partially reduce the global difference between the energy consumption of the two designs. Considering 1.80\$GJ⁻¹ and 0.70\$GJ⁻¹ as mean prices for the low pressure steam and for the cooling water, respectively ([Brüggemann & Marquardt, 2004](#)), a mean operational time

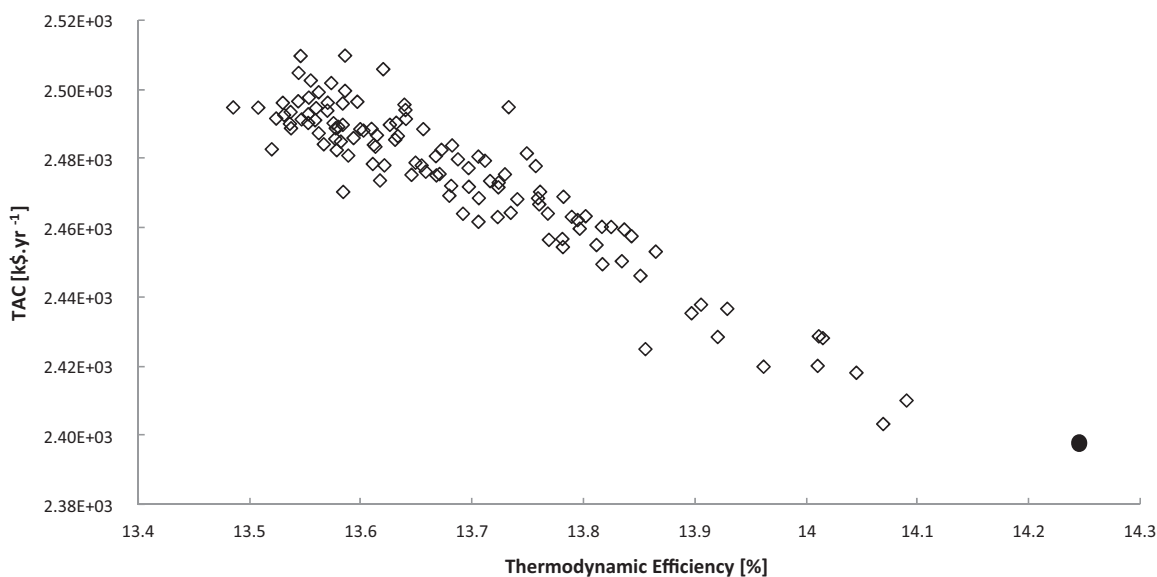


Fig. 6. Pareto front for the case study 2. The filled point corresponds to the solution compared with the SDM.

of 8000 h yr^{-1} , the corresponding operational cost is $331 \text{ k\$ yr}^{-1}$ for the SDM and $297 \text{ k\$ yr}^{-1}$ for the HMO solution. Considering a penalty of $30 \text{ k\$ yr}^{-1}$ for the HMO solution due to the higher solvent consumption, the two design options can be considered almost equivalent.

5. Conclusions

Two different column sequence design methods are presented and compared. The sequential design method (SDM) is deeply related to the synthesis procedure and, utilizing the connection between the simple column sequences and the alternative configurations included in the searching space, is possible to obtain the design parameters for the corresponding column sections through shortcut method and rigorous process simulator. On the other hand the hybrid multi-objective (HMO) algorithm has a mathematical basis and the final design is obtained exploring the variation of the parameters obtained from a preliminary short-cut method or from constraints imposed by the designer. However, it is difficult to ensure that the global optimum and not a local optimum was found. A parametric tuning procedure and different initial values must be tested in order to support the solution. As regards the two cases studies, it is possible to affirm that a local optimum was obtained.

From the cases considered it is possible to notice a general agreement between the two design methods results, even if they are based on totally different principles. This point confirms the paramount importance in developing synthesis methodologies based on the correspondence between the different types of sequences. The correspondence in column's sections among the simple column sequences and the alternatives considered used in the SDM, helps the designer to initialize the calculations reducing the efforts to define the optimal parameters. On the other hand the SDM has the drawback to directly transfer any potential error from the simple column design to the new configurations. This possibility cannot occur in HMO method since each configuration is designed independently without considering any connection with the subspace of simple column sequences. Usually the HMO method is initialized by shortcut simulation or defining parameters boundaries by means of heuristic rules. Although the agreement in the results, the time requested to get the final solution, represents a difference between the methods. Generally the HMO algorithm requires more time depending on the number of parameters to be

optimized, the number of objective functions and their interaction. For the case study 1, where 7 parameters are optimized, more than half a day is required. For the SDM the time mainly depends on the simulation abilities of the designer. For the cases examined, it is possible to estimate 6 h as a reasonable time if the more complex configurations presented for the bioethanol separation are considered.

The two methods are complementary in terms of optimal structures of the systems and optimal values of system's parameters. The sequential design method can systematically synthesize the structure of the systems and define the functionality of the components, this also the major step to approach the parameter values of system's design. On the other hand, the optimization method can search for optimal values of the design parameters and give the optimal values by multi-objective optimization satisfying two or more different criteria simultaneously. This means that it is possible to use the methods in sequence utilizing the HMO method to verify, in a small range of parameters variation, the design obtained with the SDM.

Acknowledgements

Gratefully acknowledges Sardinia Regional Government for the financial support (P.O.R. Sardegna F.S.E. Operational Program of the Autonomous Region of Sardinia, European Social Fund 2007–2013 – Axis IV Human Resources, Objective I.3, Line of Activity I.3.1 “Avviso di chiamata per il finanziamento di Assegni di Ricerca”).

References

- Ali, M. M., & Torn, A. (2004). Population set-based global optimization algorithms: Some modifications and numerical studies. *Computers and Operations Research*, 31, 1703–1725.
- Aviles Martinez, A., Saucedo-Luna, J., Segovia-Hernandez, J. G., Hernandez, S., Gomez-Castro, F. I., & Castro-Montoya, A. J. (2012). Dehydration of bioethanol by hybrid process liquid–liquid extraction/extractive distillation. *Industrial and Chemical Engineering*, 51, 5847–5855.
- Brüggemann, S., & Marquardt, W. (2004). Rapid screening of design alternatives for nonideal multiproduct distillation processes. *Computers and Chemical Engineering*, 29, 165–179.
- Caballero, J. A., & Grossmann, I. E. (2004). Design of distillation sequences: From conventional to fully thermally coupled distillation systems. *Computers and Chemical Engineering*, 28, 2307–2329.
- Errico, M., & Rong, B.-G. (2012a). Modified simple column configurations for quaternary distillations. *Computers and Chemical Engineering*, 36, 160–173.

- Errico, M., & Rong, B.-G. (2012b). Synthesis of new separation processes for bioethanol production by extractive distillation. *Separation and Purification Technology*, 96, 58–67.
- Errico, M., Rong, B.-G., Tola, G., & Spano, M. (2013). Optimal synthesis of distillation systems for bioethanol separation. Part 2. Extractive distillation with complex columns. *Industrial and Engineering Chemistry*, 52, 1620–1926.
- Errico, M., Rong, B.-G., Tola, G., & Turunen, I. (2009). Synthesis of multicomponent distillation systems with less than $n - 1$ columns. *Chemical Engineering and Processing*, 48, 907–920.
- García-Herreros, P., Gomez, J. M., Gil, I. D., & Rodriguez, G. (2011). Optimization of the design and operation of an extractive distillation system for the production of fuel grade ethanol using glycerol as entrainer. *Industrial and Engineering Chemistry Research*, 50, 3977–3985.
- Giridhar, A., & Agrawal, R. (2010). Synthesis of distillation configurations: I. Characteristics of a good searching space. *Computers and Chemical Engineering*, 34, 73–83.
- Golberg, D. E. (1989). *Genetic algorithms in search, optimization, and machine learning*. USA: Addison-Wesley.
- Hohmann, E. C., Sander, M. T., & Dunford, H. A. (1982). A new approach to the synthesis of multicomponent separation schemes. *Chemical Engineering Communications*, 17, 273–284.
- Kim, J. K., & Wankat, P. C. (2004). Quaternary distillation systems with less than $N - 1$ columns. *Industrial and Engineering Chemistry Research*, 43, 3838–3846.
- Kennedy, J., & Eberhart, R. (1995). Particle swarm optimization. *Proceedings of IEEE international conference on neural networks*, 4, 1942–1948.
- Martínez-Iranzo, M., Herrero, J. M., Sanchis, J., Blasco, X., & García-Nieto, S. (2009). Applied Pareto multi-objective optimization by stochastic solvers. *Engineering Applications of Artificial Intelligence*, 22, 455–465.
- Panduco, M. A., & Brizuela, C. A. (2009). A comparison of genetic algorithms, particle swarm optimization and the differential evolution method for the design of scannable circular antenna arrays. *Progress in Electromagnetics Research B*, 13, 171–186.
- Rangaiah, G. P. (2010). *Stochastic global optimization techniques and applications in chemical engineering: Techniques and applications in chemical engineering*. Singapore: World Scientific Publishing Co. Pte. Ltd.
- Rong, B.-G., & Errico, M. (2012). Synthesis of intensified simple column configurations for multicomponent distillations. *Chemical Engineering and Processing*, 62, 1–17.
- Sacco, W. F., Henderson, N., Rios-Coelho, A. C., Ali, M. M., & Pereira, C. M. N. A. (2009). Differential evolution algorithms applied to nuclear reactor core design. *Annals of Nuclear Energy*, 36, 1093–1099.
- Seader, J. D., & Westerberg, A. W. (1977). A combined heuristic and evolutionary strategy for synthesis of simple separation sequences. *AIChE Journal*, 23, 951–954.
- Seider, W. D., Seader, J. D., Lewin, D. R., & Widagdo, S. (2009). *Product and process design principles. synthesis, analysis and evaluation*. John Wiley and Sons, Inc.
- Shah, V. H., & Agrawal, R. (2010). A matrix method for multicomponent distillation sequences. *AIChE Journal*, 56, 1759–1775.
- Sharma, S., & Rangaiah, G. P. (2010). A hybrid multi-objective optimization algorithm. In *Proceedings of the international symposium on design, operation and control of chemical processes* (pp. 1494–1504).
- Shenvi, A. A., Shah, V. H., Zeller, J. A., & Agrawal, R. (2012). A synthesis method for multicomponent distillation sequences with fewer columns. *AIChE Journal*, 58, 2479–2494.
- Storn, R. M., & Price, K. V. (1997). Differential evolutions a simple and efficient heuristic for global optimization. *Journal of Global Optimization*, 11, 341–359.
- Taylor, M., & Wankat, P. C. (2004). Increasing the energy efficiency of extractive distillation. *Separation Science and Technology*, 39, 1–17.
- Thompson, R. W., & King, C. J. (1972). Systematic synthesis of separation schemes. *AIChE Journal*, 18, 941–948.
- Turton, R., Bailie, R. C., Whiting, W. B., & Shaeiwitz, J. A. (2004). *Analysis, synthesis and design of chemical process* (2nd ed.). USA: Prentice Hall.