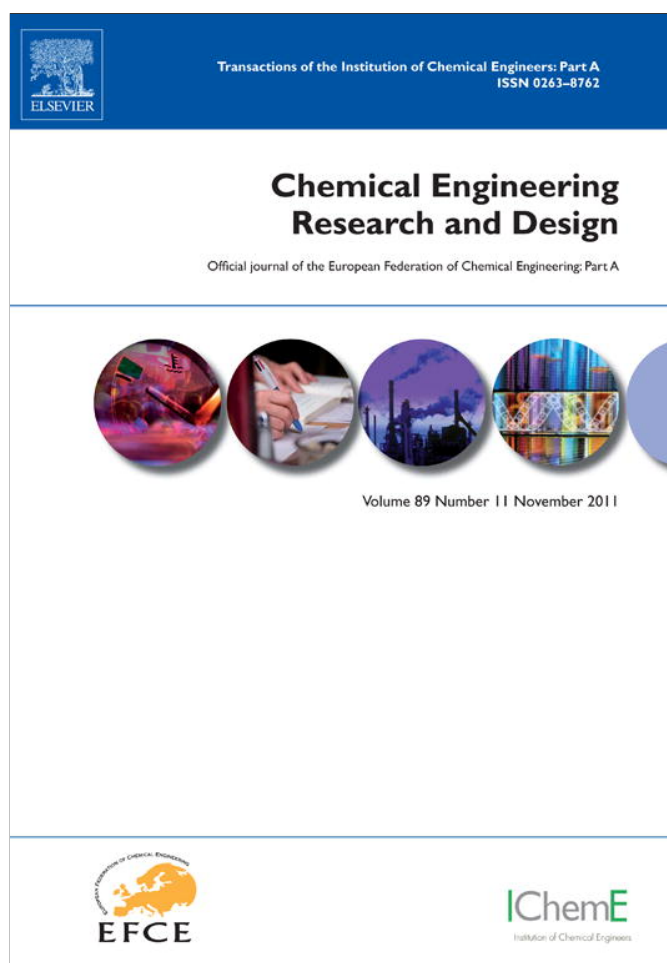


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Analysis of control properties of intensified distillation sequences: Reactive and extractive cases

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ABSTRACT

In this work, we obtain and compare the control properties of thermally coupled reactive distillation sequences and thermally coupled extractive distillation sequences with those of conventional reactive and extractive distillation configurations. All sequences have been designed using a multiobjective genetic algorithm with restrictions. The theoretical control properties of those schemes were obtained using the singular value decomposition technique in all frequency domain. In order to complete the control study, the distillation options were subjected to closed-loop dynamic simulations. The effects of total stages, reactive stages, and extractant/feed ratio on the energy consumption and control properties are obtained for the intensified distillation options. The results show that there are cases in which integrated reactive and extractive sequences do not only provide significant energy savings with respect to the conventional reactive and extractive arrangements, but also may offer dynamic advantages in high energy consumption conditions.

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Keywords: Process intensification; Thermally coupling; Pareto front; Control properties; Extractive distillation; Reactive distillation

1. Introduction

Process intensification (PI) concerns the design of novel equipment based on scientific principles and new production methods and is obtained in using either multifunctional equipments, or new operating modes, or microengineering and microtechnology for both high throughput and formulation screening, and for chemical production. Thus process intensification leads to more or less complex technologies that replace large, expensive, energy-intensive equipment or processes with ones that are smaller, less costly, more efficient plants, minimizing environmental impact, increasing safety and improving remote control and automation, or that combine multiple operations into a single apparatus or into fewer devices (Charpentier, 2007). Economic and environmental considerations have encouraged industry to focus on technologies based on process intensification. Reactive and extractive distillations are excellent examples of process intensification. The intensification of reaction or extraction and distillation

in one unit may yield several advantages: (1) combination of the reaction or extraction and separation into one unit leads to significant capital saving; (2) purity or conversion can be improved by continuous removal of products from reaction or extractive zone; and (3) direct heat integration decreases the heat demand. Additionally, motivated by the large energy requirements of distillation, researchers have developed several column arrangements that can bring savings in both energy and capital cost. Reported studies reveal that the thermally coupled distillation system (TCDS) provides the energy reduction in distillation columns (Triantafyllou and Smith, 1992; Hernández and Jiménez, 1996, 1999; Grossmann et al., 1999; Kim, 2000; Amminudin et al., 2001; Muralikrishna et al., 2002; Kim et al., 2002, 2003; Segovia-Hernández et al., 2006; Lee et al., 2008). Also TCDS are an example of process intensification: for example, Premkumar and Rangaiah (2009) showed that retrofitting the existing 2-conventional column distillation systems to dividing wall columns is very attractive both economically and for its reduced energy requirements. TCDS

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have been successfully used in the chemical industry for the separation of hydrocarbon mixtures, and recent applications include the separation of azeotropic mixtures and reactive systems (Hernández et al., 2009; Gutiérrez-Guerra et al., 2009). These latest applications are the most representative cases of process intensification because it integrates reaction or extraction and separation in the same unit, and results in energy requirement savings that can be translated into reductions of total annual cost and miniaturization of the schemes.

Design issues for reactive and extractive distillation systems are significantly more complex than those involved in ordinary distillation. Several optimization methods that utilize mathematical programming models for the design of reactive and extractive distillation columns have been proposed (Jackson and Grossmann, 2001; Farkas et al., 2008). These methods are able to achieve the global minimum on energy consumption, but they demand high mathematical efforts. Furthermore, the formulation of such models is difficult and time-consuming. Due to the non-monotonic and local optimum properties of the search space, conventional derivative-based optimization algorithms turned out to be incapable of finding the global optimum design in most cases. Stochastic optimization algorithms are capable of solving, robustly and efficiently, the challenging multi-modal optimization problem, and they appear to be a suitable alternative for the design and optimization of complex separation schemes. Among stochastic algorithms, genetic algorithms (GA) have shown their merits in large-scale parallelism search, approaching the global optimum quickly and steadily. Genetic algorithms have several features that make them attractive for solving optimization problems with modular simulators, where the model of each unit is only available in an implicit form (Bravo-Bravo et al., 2010; Vázquez-Castillo et al., 2009, among others).

Contrary to the conventional distillation process, the dynamic of the extractive and reactive columns have been little explored in the published literature, although some authors have attacked this problem. Recently, Luyben (2008a,b,c) has studied the dynamic performance of the conventional extractive distillation sequences. Dynamic simulation results show that all extractive systems are controllable. Ibarra-Sánchez and Segovia-Hernández (2010) analyze the control properties of thermally coupled extractive distillation schemes. The results showed that the energy savings predicted in the complex extractive distillation sequence can be achieved along with good dynamic behavior and reductions in greenhouse gas emissions. In the case of reactive distillation, Huang and Wang (2007) have studied a reactive distillation column for decomposing methyl tertiary butyl ether into isobutylene and methanol. Process dynamics of the resultant process design were further investigated, and a noticeable improvement was gained in process controllability. Luyben and Yu (2008d) have presented an extensive study on the control properties reactive distillation systems for different case studies. Hernández et al. (2009) have studied the control properties in a thermally coupled reactive distillation sequence for the production of ethyl acetate. The results showed that the energy savings predicted in the intensified reactive distillation sequence can be achieved with good dynamic behavior.

In this work, we analyze the control properties of thermally coupled reactive distillation sequences and thermally coupled extractive distillation sequences in comparison with conventional reactive and extractive distillation configurations. We analyzed two cases of study: (1) the production of biodiesel

using reactive distillation (Fig. 1). Particularly, in this paper the esterification of methanol and lauric acid is studied. Fatty esters are important fine chemicals used in the manufacturing of cosmetics, detergents, and surfactants. Particularly, methyl ester could play a significant role in the future as a major component of biodiesel fuels. (2) The extractive distillation of a mixture of ethanol–water using as entrainer ethylene glycol (Fig. 2). All intensified systems were compared with conventional configurations. All sequences have been designed using a multiobjective genetic algorithm with restrictions. We study the theoretical control properties of those schemes using the singular value decomposition technique in all frequency domain and we also present an analysis on the closed loop behavior with dynamic rigorous simulations.

2. Design and optimization strategy of process intensified distillation systems

As a design tool for conventional and intensified schemes, we use a multiobjective genetic algorithm with constraints, which is coupled to Aspen Plus simulator. Multiobjective optimization has found numerous applications in Chemical Engineering due to achieve sustainable processes and manufacturing (Rangaiah, 2010). 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. The set of solutions integrates the Pareto front (Gutiérrez-Antonio and Briones-Ramírez, 2009). This set of optimal solutions allows the engineer to choose a good compromise between several goals, by picking a point somewhere along the Pareto front. Once that Pareto fronts are obtained, an analysis is realized on the resulting data, looking for some tendencies in the variables of interest of these sequences, and also to find the best scheme for this particular reaction.

For the conventional and intensified distillation columns, the objectives of the optimization problem include minimization of total number of stages, reactive stages, the extracting agent flow, and the heat duty of the sequence, but constrained by the desired purities and recoveries:

$$\text{Min}(Q_i, \text{IAE}_m, \text{TC}) = f(R_i, N_i, N_{F,i}, F_{EA}, F_k, N_k, N_{r1}, N_{r2}, F_{PS,o}) \quad (1)$$

Subject to

$$\bar{y}_m \geq \bar{x}_m$$

where R_i is the reflux ratio, $N_{F,i}$ is the number of the feed stage and N_i is the number of stages of column i of the sequence, Q_i is the reboiler duty of column i of the sequence, IAE_m is the integral of the absolute error in the controller m , TC is the total cost of the sequence, F_{EA} is the extracting agent flow, N_{r1} and N_{r2} are the initial and final stages of the reactive section N_R in column j , F_k and N_k are the value and location of the interconnection flow k . Also, the product stream flows, $F_{PS,o}$, are manipulated due to this also being required to manage the recoveries of the components along with their purities; y_m and x_m are the vectors of obtained and required purities and recoveries for the m components, respectively. In each column there are three objectives to minimize: IAE , TC and Q . For these sequences the objectives are in competition, so they have to be optimized simultaneously. The manipu-

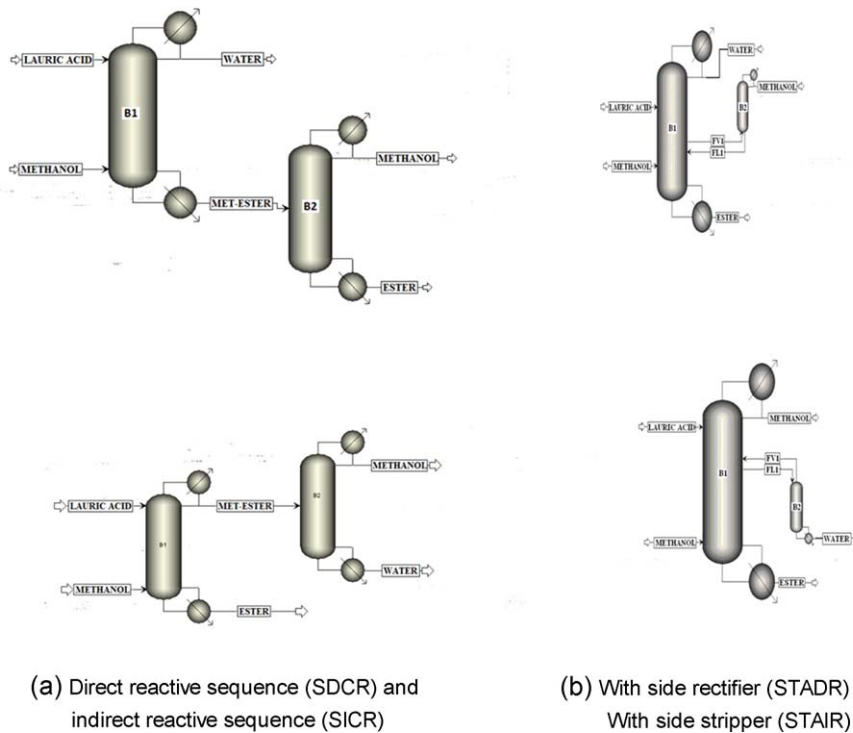


Fig. 1 – Process for the production of biodiesel using conventional schemes and configuration with thermally coupling with a side column.

lated variables include reflux ratio, total number of stages, the stage number and value of liquid and vapor interconnection flows, product streams flows, and extracting agent flow. The use of the multiobjective genetic algorithm with

constraints allows obtaining the rigorous Pareto front of the conventional and thermally coupled distillation systems: a set of non-dominated, optimal, and rigorous designs that satisfied the purities required. The term “non-dominated” means

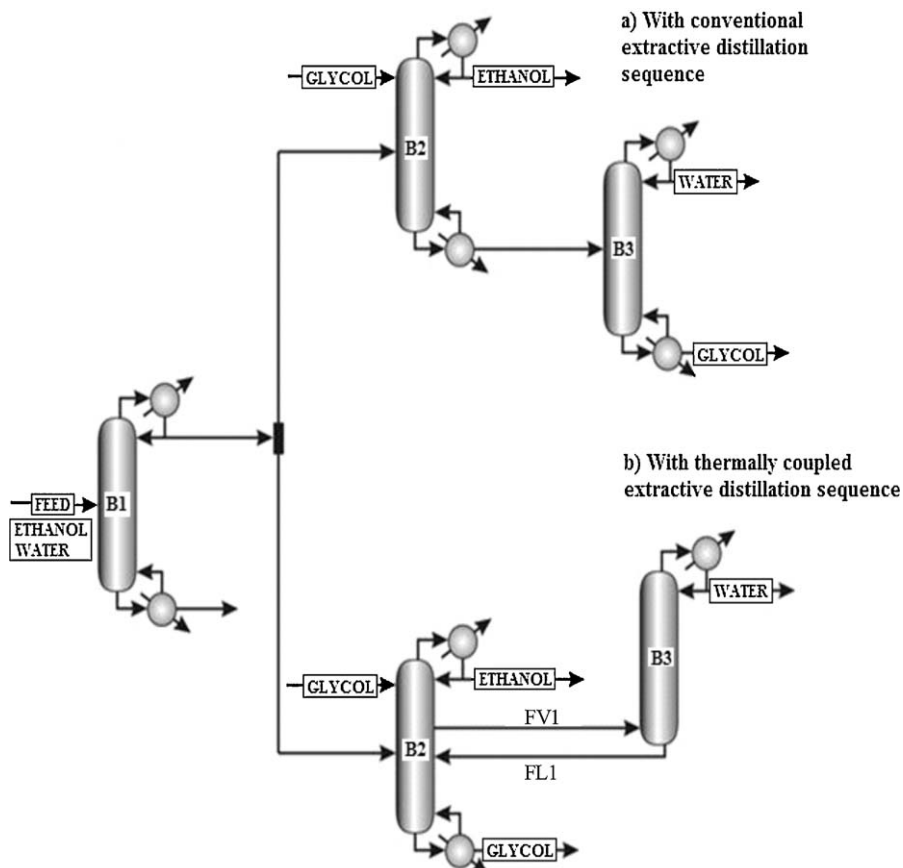


Fig. 2 – Process for the purification of bioethanol using schemes with: (a) conventional extractive distillation structure (CEDS) and (b) thermally coupled extractive distillation sequence (TCEDS).

that there is no other design that can improve one objective without worsening another one. The term “rigorous” means that all designs presented were obtained considering the complete set of MESH equations along with the phase equilibrium calculations, using the Radfrac module of Aspen Plus. The multiobjective genetic algorithm works as follows: For each run, a feasible initial design of the conventional or complex sequence is given as initial solution to the algorithm. This initial design can be obtained using the shortcut method of Fenske–Undewood–Gilliland used for the initial tray structure of thermally coupled distillation sequences in [Hernández and Jiménez \(1996, 1999\)](#). This initial design is needed as the Aspen Plus simulator requires design specifications (number of stages, feed stage, reactive stages, etc.) to start running. Additionally, a design obtained by short cut methods helps the optimizer to converge more easily and less time is required to reach the optimal solution. Starting from the initial design, the algorithm generates N individuals (i.e., new designs) to make up the initial population. The manipulated variables of each of the N individuals are sent to Aspen Plus to perform the simulation; then, the algorithm retrieves, from Aspen Plus, the values of objective functions and constraints for each individual.

It is worth of mention that in the case reactive the conversion of the reaction is not an optimization objective itself; however, we specify the recoveries of the unreacted compound and of the principal product as constraints of the problem. Thereby, indirectly the conversion is considered as an objective.

For the optimization of intensified distillation sequences, we used 2000 individuals and 40 generations as parameters of the multiobjective genetic algorithm with 0.80 and 0.05 for crossover and mutation fraction. These parameters were obtained through a tuning process. The manipulated variables are just limited to have positive values, and practically there are not restricted to certain range of values; the aim is not imposed artificial limits to them. The time employed for the optimization of each sequence is between 8 and 10 h on a Xeon 5410 workstation at 2.33 GHz with 8 GB of RAM. Of the total computational time required, 95% is consumed by the simulations performed in Aspen Plus; in other words, the most time-consuming activity is the evaluation of objective and constraint functions. For more detailed information about this algorithm and its link to Aspen Plus, the reader is referred to the original work ([Gutiérrez-Antonio and Briones-Ramírez, 2009](#)).

3. Control analysis

The control analysis was conducted in two parts. First, the singular value decomposition (SVD) technique was used to obtain a comparative outlook on the theoretical control properties of the seven distillation structures. Then, closed loop control tests were run to complement the theoretical control properties predicted by SVD. The closed loop runs were conducted under servo and load disturbance scenarios using proportional-integral (PI) controllers. While several types of controllers have been studied in the literature of reactive and extractive distillation, PI controllers ranging from simple to complex nonlinear model predictive controllers, in industrial applications the PI controllers are still widely used because of its simplicity and effectiveness ([Luyben and Yu, 2008d](#)). For instance in a recently paper published by [Baghmisheh et al.](#)

(2010), it has been successfully used PI controllers for temperature control of complex quaternary distillation sequences. The aim of the control analysis is to identify the best structures from a dynamic point of view, and to corroborate if the intensified arrangements indeed improve the dynamic characteristics of the conventional configuration.

3.1. Open loop analysis

Open loop dynamic responses to changes around the assumed operating point (for each configuration) were obtained. The magnitude of step changes made to obtain the open-loop dynamic responses is 0.5% of the manipulated variable. The responses were obtained through the use of Aspen Dynamics. Transfer function matrices (G) were then collected for each case, and they were subjected to singular value decomposition (SVD):

$$G = V\Sigma W^H \quad (2)$$

where $\Sigma = \text{diag}(\sigma_1, \dots, \sigma_n)$, $\sigma_i =$ singular value of $G = \lambda_i^{1/2}(GG^H)$, $V = (v_1, v_2, \dots)$ matrix of left singular vectors, and $W = (w_1, w_2, \dots)$ matrix of right singular vectors. Two parameters of interest are the minimum singular value σ^* and the ratio maximum to minimum singular values, or condition number

$$\gamma = \frac{\sigma^*}{\sigma_*} \quad (3)$$

The minimum singular value is a measure of the invertibility of the system and represents a measure of the potential problems of the system under feedback control. The condition number reflects the sensitivity of the system under uncertainties in process parameters and modeling errors. These parameters provide a qualitative assessment of the theoretical control properties of the alternate designs. The systems with higher minimum singular values and lower condition numbers are expected to show the best dynamic performance under feedback control ([Gabor and Mizsey, 2008](#)). In this case we cover a sufficiently complete range of frequencies. Similar studies have been reported by [Jantes-Jaramillo et al. \(2008\)](#) and [Gómez-Castro et al. \(2008\)](#), among others, for control analysis in studies of thermally coupled distillation systems.

3.2. Closed loop analysis

One of the key parts for the dynamic analysis is the selection of control outputs and manipulated variables for each control loop. Although more formal techniques to define the control loops for the complex columns may be used (for instance the relative gain array method), we based our selection on practical considerations. For example, for any sequence, the control of the lightest component was manipulated with the top reflux flowrate. The closed loop analysis was based on PI controllers. Several alternatives exist for tuning up the controller parameters. We attempted a common ground for comparison by optimizing the controller parameters, proportional gains (K_C) and reset times (τ_i), for each conventional and integrated scheme following the integral of the absolute error (IAE) criterion. For the integrated arrangements, the procedure is particularly complicated because of the interactions of the multivariable control problem. For the integrated arrangements, the procedure is particularly complicated because of the interactions of the multivariable control problem. For

Table 1 – Kinetic parameters for biodiesel reaction.

Reaction	i	k_i^0 (mol g ⁻¹ s ⁻¹)	$E_{A,i}$ (kJ mol ⁻¹)
Esterification	1	9.1164×10^5	68.71
Hydrolysis	-1	1.4998×10^4	64.66

these cases, the tuning procedure was conducted taking one control loop at a time; the parameters thus obtained were taken for the following control loop until the three loops were considered.

4. Case of study

For the production of biodiesel the systems include two feed streams; the first is lauric acid with a flow of 45.4 kmol/h as saturated liquid at 1.5 bar, and the second is methanol with a flow of 54.48 kmol/h as saturated vapor at 1.5 bar. The design objective is a process for high-purity fatty ester, over 99.9% mass fraction. It is important to highlight that this equilibrium reaction is usually catalyzed using sulfuric acid or p-toluensulfonic acid. The kinetic model reported in Steinigeweg and Gmehling (2003) was used (Table 1). For this class of reactive systems, thermodynamic models such as UNIFAC can be used to calculate vapor–liquid or vapor–liquid–liquid equilibrium. For the case of purification of bioethanol, a dilute feed of 100 lb-mol/h ethanol in water [10% in moles of ethanol in water. This is a typical yield for fermentation of sugarcane bagasse (Kummar et al., 2009)] as saturated liquid at 1 atm is introduced into a conventional distillation column that removes the binary homogeneous azeotrope as distillate. This study focuses on the separation stage for ethanol with a high mass fraction (0.995). The bottoms product of the first distillation column is almost pure water. This conventional distillation column is needed in all three distillation options. The first alternative (Fig. 2a) uses an extractive conventional distillation column with ethylene glycol as entrainer; the distillate of the column is ethanol with a mass fraction of 0.995. The second option (Fig. 2b), in the extractive stage of the separation, uses a thermally coupled extractive distillation scheme. Distillation columns coupled to side columns and the Petlyuk distillation column are the most representative options among thermally coupled distillation sequences (Dejanovic et al., 2010). The UNIQUAC model was used to predict thermodynamic properties.

5. Results

In this section, we present the set of optimal designs, called Pareto front, for the conventional and thermally coupled reactive and extractive distillation sequences. Fig. 3 shows the energy consumption of the optimal designs for conventional and thermally coupled reactive sequences. It is clear that the thermally coupled reactive sequences have lower energy consumptions; however, it is important to remark that these savings are not obtained by an increase in the total number of stages of the sequence. In the case of thermally coupled reactive configuration, the production of ester requires less energy because it is separated from the mixture of unreacted methanol, present in a lower proportion, and water. So, being the ester the more abundant component, the separation task is easier and the energy requirements are lower. On the other hand, in the indirect reactive sequences, both conventional and thermally coupled, the production of ester requires more energy because it is separated from the mixture of water and unreacted methanol, which is present in a higher proportion since they are in the first column. So, being the ester the less abundant component and its purity specification very high, the separation is harder and the energy requirements are greater. These findings are important because, depending on the type of reaction and byproducts involved, we can know the better sequence beforehand. For instance, from this analysis we can expect that then STAIR sequence could be a good option, in terms of energy requirements, for reactions where the principal product is always more abundant than the unreacted component and byproducts.

Similarly, in the case of extractive configurations, the results in the Pareto front can be summarized as follows: (i) reducing solvent feed ratio of the complex extractive distillation systems causes a reduction of energy savings in comparison with the conventional distillation sequence and consequently the total annual cost will be increased; and (ii) the energy savings achieved by complex extractive schemes are in the range between 20 and 30% in contrast to the conventional arrangement (see Tables 2 and 3). It is also clear that reactive designs with minimal energy consumption have a higher number of stages, in comparison with those configurations that show high energy consumption. For all designs can be seen that the values of reflux ratio are very similar regardless of the amount of energy consumed in the reboiler. In all cases it was observed that biodiesel reaches a value of 0.999 in mass purity. A common characteristic is that the reactive

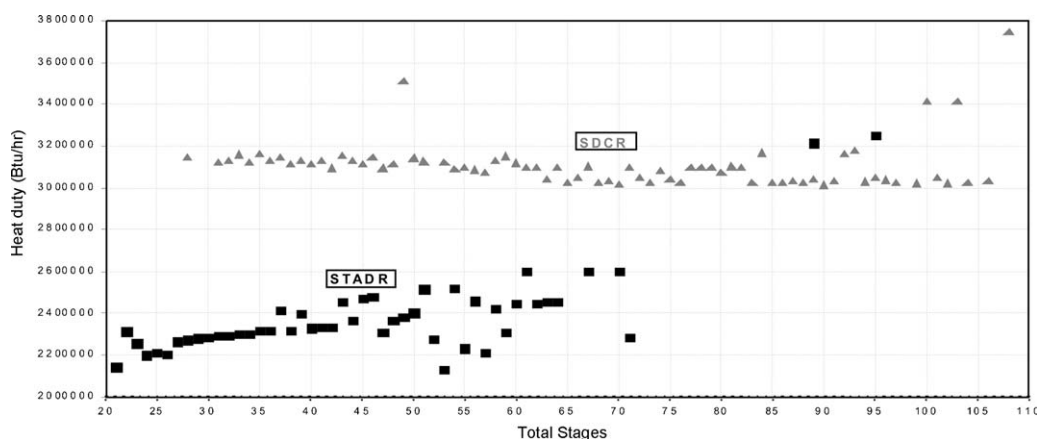


Fig. 3 – Total number of stages versus energy consumption of thermally coupled (STADR) and conventional reactive direct sequences (SDCR).

Table 2 – Selected designs from Pareto fronts for reactive systems.

Design parameters	SDCR 14 (minimum IAE and minimum consumption design)	SIGR 710 (minimum design)	SIGR 131 (minimum energy consumption design)	STADR 21 (minimum design)	STADR 9 (minimum energy consumption design)	STAIR 16 (minimum design)	STAIR 43 (minimum energy consumption design)
Reflux ratio column B1	2.61	2.38	2.3	2.63	2.58	19.2129445	18.8829445
Reflux ratio column B2	0.052524246	2.27450690	1.410519382	0.06255198	0.04605535	0	0
Number of stages of column B1	72	10	10	32	41	41	23
Number of stages of column B2	18	26	62	5	7	22	3
Number of reactive stages	30	8	8	30	31	23	18
Total number of stages	90	36	72	37	48	63	26
Feed stream flow of lauric acid (lb-mol/h)	100	100	100	100	100	100	100
Feed stream flow of methanol (lb-mol/h)	120	120	120	120	120	120	120
Total feed stream flow (lb-mol/h)	220	220	220	220	220	220	220
Heat duty of column B1 (BTU/h)	110010.336	3733029.68	3264903.03	3214696.93	3031432.15	2223776.04	2084224.35
Heat duty of column B2 (BTU/h)	2902986.59	1068276.66	813116.379	0	0	306099.532	286986.377
Total heat duty (BTU/h)	3012996.93	4801306.34	4078019.41	3214696.93	3031432.15	2529875.57	2371210.73
Stages of interconnection flow FV1	NA	NA	NA	11	11	8	7
Stages of interconnection flow FL1	NA	NA	NA	4	4	7	7
Interconnection liquid flow FL1 (lb-mol/h)	NA	NA	NA	0.18263521	0.18263521	116.770762	116.770762
Interconnection vapor flow FV1 (lb-mol/h)	NA	NA	NA	19.3754263	19.3754263	16.7561426	16.7561426
Stage of the side stream	NA	NA	NA	1	1	22	3
Operating pressure of column B1 (psi)	14.7	14.7	14.7	14.7	14.7	14.7	14.7
Operating pressure of column B2 (psi)	14.7	14.7	14.7	14.7	14.7	14.7	14.7
Purity of biodiesel recovered (%)	0.999	0.999	0.999	0.999	0.999	0.999	0.999
Purity of methanol recovered (%)	0.98	0.98	0.98	0.98	0.98	0.98	0.98
Purity of water recovered (%)	0.98	0.98	0.98	0.98	0.98	0.98	0.98
Thermodynamic efficiency (%)	30.52949545	21.39871689	24.53163206	32.50872096	18.38704762	47.32993846	47.61364819
CO ₂ emission (tonne/year)	2301.396548	3466.400205	2960.695251	2528.404529	2353.87716	1936.669009	1799.642648
Total annual cost (\$/year)	1923890.23	1152447.46	1665430.788	986498.50	1317329.44	1191148.219	793723.2346

Table 3 – Selected designs from Pareto fronts for extractive systems.

Design parameters	CEDS (minimum IAE design)	CEDS (minimum energy consumption design)	TCEDS (minimum IAE design)	TCEDS (minimum energy consumption design)
Reflux ratio column B1	0.570	0.44908356	0.170	0.165
Reflux ratio column B2	1.110	1.16653916	1.457	1.29
Reflux ratio column B3	0.270	0.27837878	0.275	0.06909918
Number of stages of column B1	11	10	21	20
Number of stages of column B2	27	38	22	28
Number of stages of column B3	7	9	6	5
Total number of stages	45	57	49	53
Stage of the binary mixture	3	3	10	11
Stage of the extractive agent	5	5	3	3
Feed stream flow of ethanol (lb-mol/h)	10	10	10	10
Feed stream flow of water (lb-mol/h)	90	90	90	90
Total feed stream flow (lb-mol/h)	100	100	100	100
Extractive agent flow ethylene glycol (lb-mol/h)	8.796424	8.03003855	23.4466911	14.0540982
Total heat duty (Btu/h)	1106191.68	1073656.15	1180005.96	1084790.36
Ethanol recovered (lb-mol/h)	9.999057	9.98730264	9.917457	9.95659365
Water recovered (lb-mol/h)	89.8777503	89.9371429	89.7745876	89.8750259
Ethylene glycol recovered (lb-mol/h)	8.752836	7.98956741	23.38	14.0423906
Stage of the interconnection vapor flow FV1	NA	NA	19	23
Stage of the interconnection liquid flow FL1	NA	NA	11	10
Interconnection liquid flow FL1 (lb-mol/h)	NA	NA	3	3
Interconnection vapor flow FV1 (lb-mol/h)	NA	NA	5	5
Operating pressure of column B1 (psi)	14.7	14.7	14.7	14.7
Operating pressure of column B2 (psi)	14.7	14.7	59.7	59.7
Operating pressure of column B3 (psi)	24.7	24.7	39.7	39.7
Purity of the ethanol recovered (%)	0.9926	0.99378389	0.9910	0.99222934
Purity of the water recovered (%)	0.9995	0.99940834	0.9991	0.99946288
Purity of the ethylene glycol recovered (%)	0.9945	0.99994856	0.9914	0.99617234
Thermodynamic efficiency (%)	7.88	8.13	18.95	16.48
CO ₂ emissions (lb/h)	81.71	79.41	103.4	93.53
Total annual cost (\$/year)	281789	310311	299401	301457

section includes almost the entire first column. The energy consumption in the conventional indirect sequence is greater, because the reaction and separation must be performed in the first column, and the purity required for ester is very high. In the thermally coupled sequences, we can observe the opposite. The higher efficiency is observed in the indirect sequence, since an easier separation (unreacted methanol from ester) is performed in the first column; in the direct sequence, the first column performs a harder separation, so the efficiency is lower. In other words, the easier separation must be performed in the first column, and the harder in the next ones;

this observation has been observed also for non-reactive separations.

Table 3 also shows that the best extractive systems with best control properties (design with minimum IAE value) shows a smaller number of stages compared with designs with minimal energy consumption. In both cases the reflux ratios are very similar. The amount of extracting agent is higher in the coupled system compared with conventional systems.

In general, complex thermally coupled distillation sequences are more efficient and with diminution in the energy consumption because the remixing in the interme-

Table 4 – Transfer function matrices for (a) STAIR and (b) STADE with better open loop responses.

a		R_1	Q_1	Q_2	
x_A		$\frac{-5.4912}{0.0065 S^2+0.1619 S+1} + \frac{0.1892}{0.1829 S+1}$	$\frac{0.0096}{0.3583 S^2+1.1972 S+1} - \frac{0.0068}{3.0273 S+1}$	$\frac{1.2612}{0.7110 S+1}$	
x_B		$\frac{0.5032}{0.0163 S^2+0.2556 S+1} - \frac{0.1644}{0.5293 S+1}$	$\frac{0.0044}{1.0523 S+1}$	$\frac{-0.5568}{0.8880 S+1}$	
x_C		$\frac{0.2696}{0.0847 S+1}$	$\frac{-0.1852}{0.04305 S+1}$	$\frac{-0.0064}{0.0028 S^2+0.1063 S+1} + \frac{0.0308}{1.1240 S+1}$	
b		R_1	Q_1	R_2	Q_2
x_A		$\frac{0.0008}{1+14 S}$	$\frac{-0.1276}{1+2.3 S}$	$\frac{0.0008}{1+9 S}$	$\frac{0.0008}{1+9 S}$
x_B		$\frac{1.182}{1+6 S} - \frac{1.7152}{1+34 S}$	$\frac{-6.7136}{1+47 S}$	$\frac{-0.7448}{400 S^2+40 S+1}$	$\frac{-4.0756}{1+5.25 S} + \frac{3.7516}{1+22 S}$
x_C		$\frac{-0.1084}{1+5.5 S} + \frac{0.1204}{1+26 S}$	$\frac{0.2672}{1+40 S}$	$\frac{0.0348}{1+40 S}$	$\frac{0.0984}{1+3.6 S} - \frac{0.1884}{1+24 S}$
x_D		$\frac{0.2768 e^{-3s}}{400 S^2+40 S+1}$	$\frac{1.1248}{1+33 S}$	$\frac{0.118}{1+33 S}$	$\frac{0.76 e^{-3s}}{169 S^2+26 S+1}$

diante component, presented naturally in the conventional distillation sequences and translated into higher energy consumption, is reduced significantly for the presence of recycles (Triantafyllou and Smith, 1992; Hernández et al., 2003).

The theoretical control properties of thermally coupled distillation sequences were obtained using SVD technique. The dynamic responses were adjusted to transfer functions and arranged into transfer function matrices. Table 4 presents typical transfer function matrices for separation of the (a) STAIR with better open loop properties and (b) STADE with better open loop properties. The results obtained in the Pareto front were analyzed for both cases of study. To compare the controllability of the different designs, their controllability indexes are analyzed (minimum singular value and condition num-

ber). In Fig. 4, the σ_* and γ for some cases of study are showed (case reactive). There are important differences between the design operated at minimum energy consumption and the scheme operated at high total annual cost.

In the case of STADR and STAIR, when they are operated at high energy consumption, number of stages and total annual cost conditions their controllability improves (for example, design STAIR 16 is a nonoptimal design and the scheme STAIR 43 shows the minimum energy consumption for this scheme). In those high energy consumption conditions, STADR and STAIR present highest value of the minimum singular value (Fig. 4); therefore, it can be expected that coupled reactive systems exhibit better control properties than the coupled sequence, in "optimal condition" (low energy consumption, number of stages and total annual cost), under

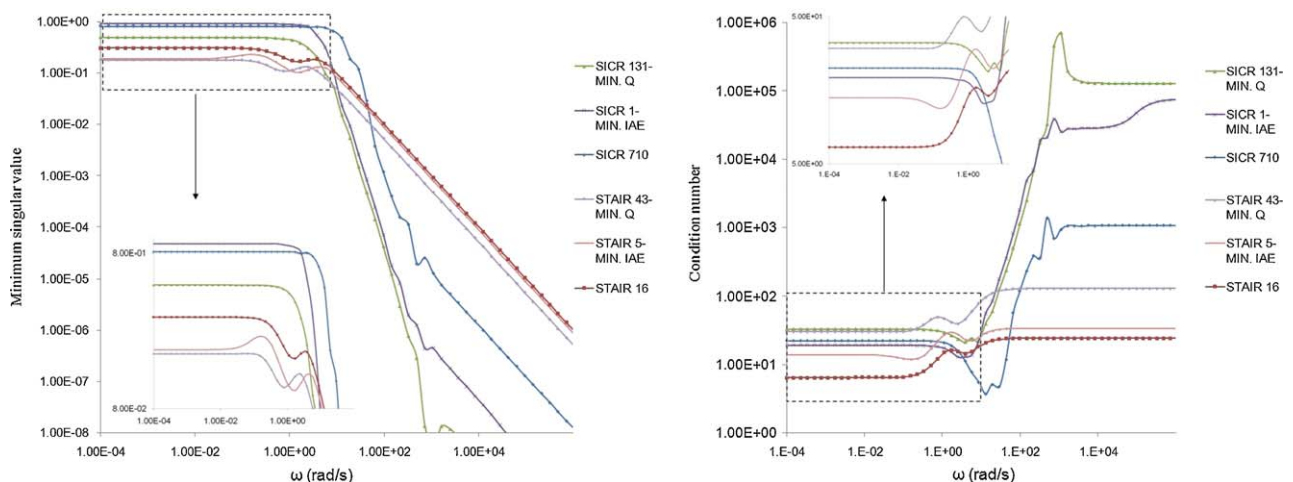


Fig. 4 – Minimum singular value and condition number for different thermally coupled reactive schemes with side stripper (STAIR) and indirect sequence (SICR).

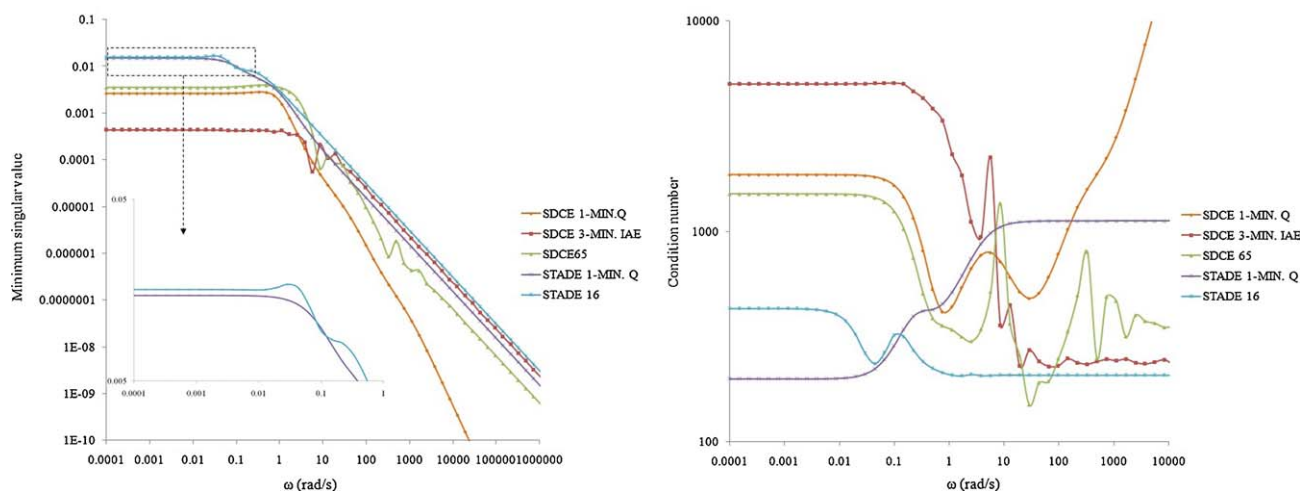


Fig. 5 – Minimum singular value and condition number for different thermally coupled extractive schemes with side rectifier (STADE) and conventional extractive sequence (SDCE).

feedback control. The results for the condition number show that coupled sequences in the high energy consumption values offer the best behavior (Fig. 4). As a result, it can be expected that thermally coupled reactive distillation system in a different operating condition is better conditioned to the effect of disturbances than the minimum energy consumption arrangement (in other words, the complex system can eliminate the disturbances better than the conventional arrangements in a high energy consumption condition). As has been explained, the operation in “nonoptimal conditions” has higher energy consumption and total annual cost than optimal conditions. Consequently when the reboiler duty, reflux ratio, number of stages and number of reactive stages are increased, the controllability has improved. The reboiler duty in the coupled systems is lower than the conventional sequence in the case when the controllability parameters are better than the optimal design.

A similar analysis using SVD was conducted for the case of extractive systems (Fig. 5). When the reboiler duty, reflux ratio, number of stages and total annual cost are increased, the controllability of the thermally coupled extractive distillation arrangement is better because the minimum singular value present highest value and the condition number show the minimum value. Also, the reboiler duty in the coupled systems is lower than the conventional sequence in high energy consumption case. As a result, it can be expected that thermally coupled extractive distillation system in a different operating condition is better conditioned to the effect of disturbances than the “optimal arrangement”. Also, the reboiler duty in the coupled extractive systems is lower than the conventional sequence in the case when the controllability parameters are better than the minimum energy consumption conditions.

The closed loop analysis should be conducted on a basis as consistent as possible. Therefore, the same type of controllers, the same tuning technique and the same control loops were used for each distillation configuration. For this initial comparative analysis, PI controllers were selected. The parameters of the controllers were tuned up through a minimization procedure of the integral of the absolute error (IAE). For each control loop and aided by Aspen Dynamics, a search procedure was conducted on the gain constant and the integral time parameters until a minimum value of the IAE was obtained. Several techniques, such the relative gain array method, can be used to fix the loops for a control system. In the case of distilla-

tion columns, however, such loops are fairly well established and used successfully in practice, at least for conventional columns. A well-known structure is based on energy balance considerations, which yields to so-called LV control structure in which the reflux flowrate L and the vapor boilup rate V (affected directly by the heat duty supplied to the reboiler) are used to control the distillate and bottom outputs compositions respectively (see for instance, Häggblom and Waller, 1992). The control loops for the complex distillation systems were chosen from extensions of the practical considerations observed for conventional distillation columns. The control objective was to preserve the output streams at their design purity specifications. For the dynamic analysis, individual set point changes for product composition were implemented for each of the three product streams. The liquid compositions for the main product streams A, B and C were taken as the controlled variables whereas, respectively, the reflux flowrate and the reboiler heat duty were chosen as the manipulated variables. For all cases (“optimal and nonoptimal conditions”), the three control loops were assumed to operate under closed loop fashion. The performance of the sequences under analysis was compared through the evaluation of IAE values for each test. Fig. 6 displays the IAE value for biodiesel stream (SDCR, SICR, STAIR and STADR schemes). The results show that the scheme in high energy consumption conditions offers the best dynamic behavior (minimum IAE value). Therefore, the results indicate that to have a major dynamic performance should be operated in a region outside the optimum of minimum energy consumption. In the case of conventional intensified reactive configurations the IAE value for all cases is higher in comparison with coupled reactive systems. This situation corroborates that operating in high energy consumption conditions is a good option. Similar results were obtained in the coupled extractive scheme: the scheme in a region outside energy consumption offers the best dynamic behavior (minimum IAE value). For the conventional and coupled extractive arrangements, Fig. 7 shows the IAE for bioethanol stream. The IAE value for all cases is higher in comparison with coupled reactive systems. In general, operating in minimum energy consumption conditions is not a good option for the case of intensified systems.

The results from the individual servo tests (a step change was induced in the set point for each product composition under single-input, single-output, SISO, feedback control at

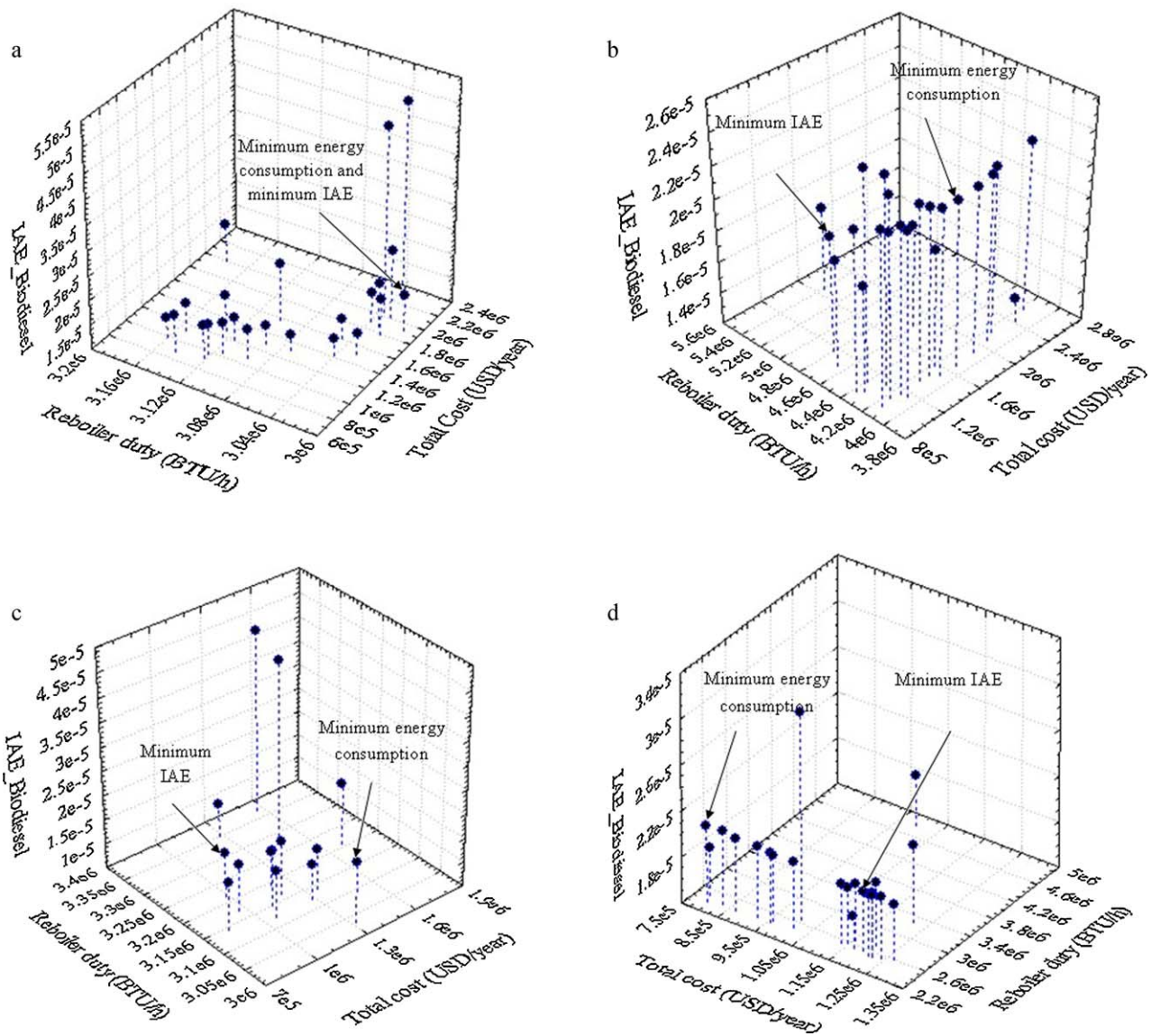


Fig. 6 – IAE value for biodiesel component in (a) SDCR, (b) SICR, (c) STADR and (d) STAIR.

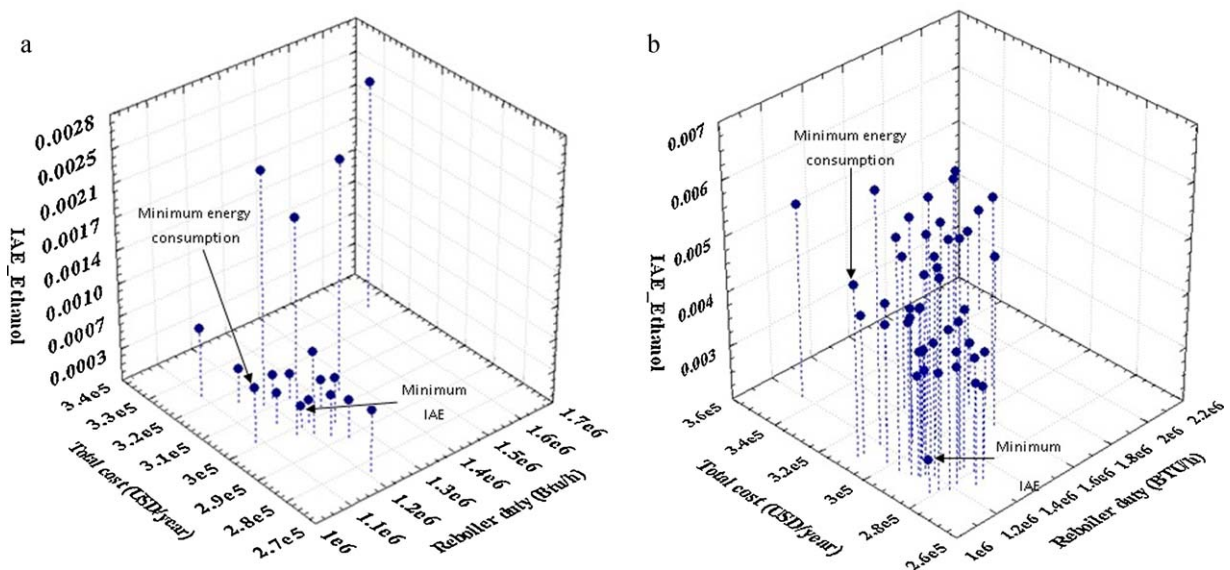


Fig. 7 – IAE value for ethanol component in (a) SDCE and (b) STADE.

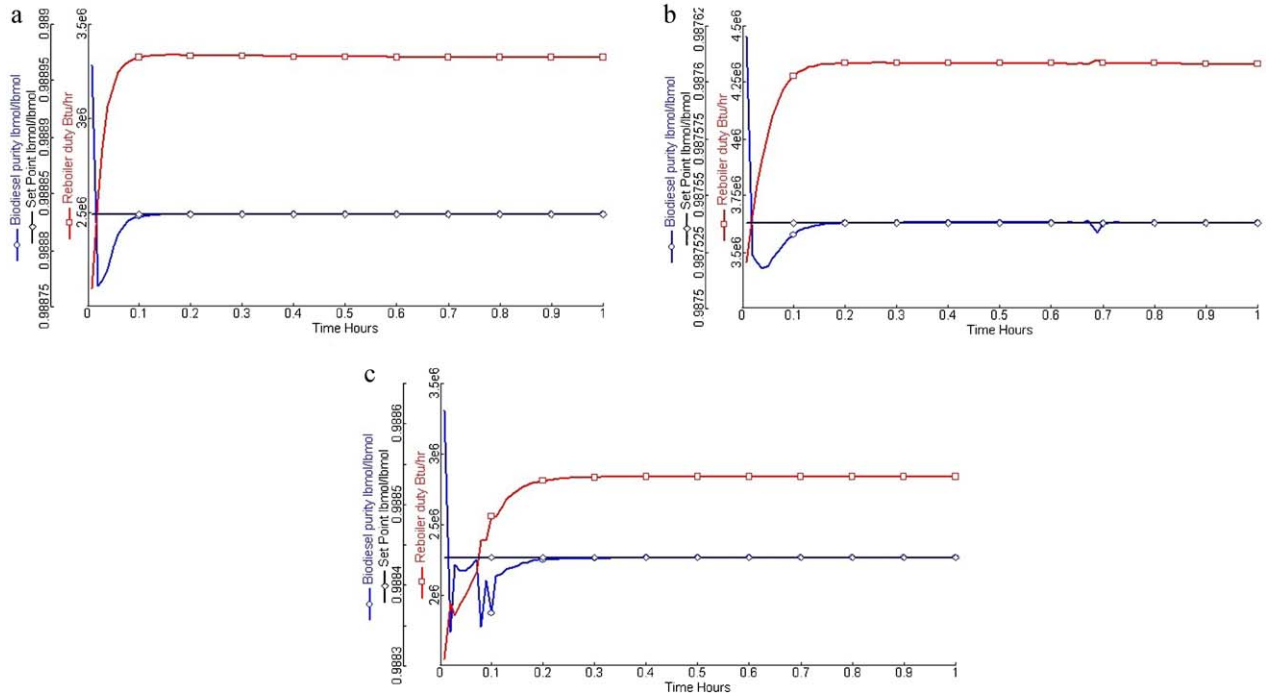


Fig. 8 – Closed loop dynamic response in SICR for biodiesel stream in the design with (a) best SVD results, (b) minimum Q and (c) minimum IAE.

each output flowrate) for some conventional and coupled reactive cases are displayed in Figs. 8 and 9. All designs are able to reach the new values of product compositions imposed as step changes. One may notice that intensified configurations in “non optimal conditions” (with side rectifier and stripper) provide the minimum settling time for the SISO control of the biodiesel component. The conventional systems show high settling times. These results are consistent with the predictions given by SVD. For the extractive system, the inten-

sified extractive configuration (Figs. 10 and 11) provides fairly responses with a minimum settling time, for “non optimal conditions”, for bioethanol stream. The conventional extractive system shows the worst response with high settling times. Those results are consistent with the predictions given by SVD.

In summary, it is clear that the reactive configurations with minimum energy consumption show a worse dynamic behavior in comparison with designs in “non optimal condition”. Similar results are showed for the case of extractive

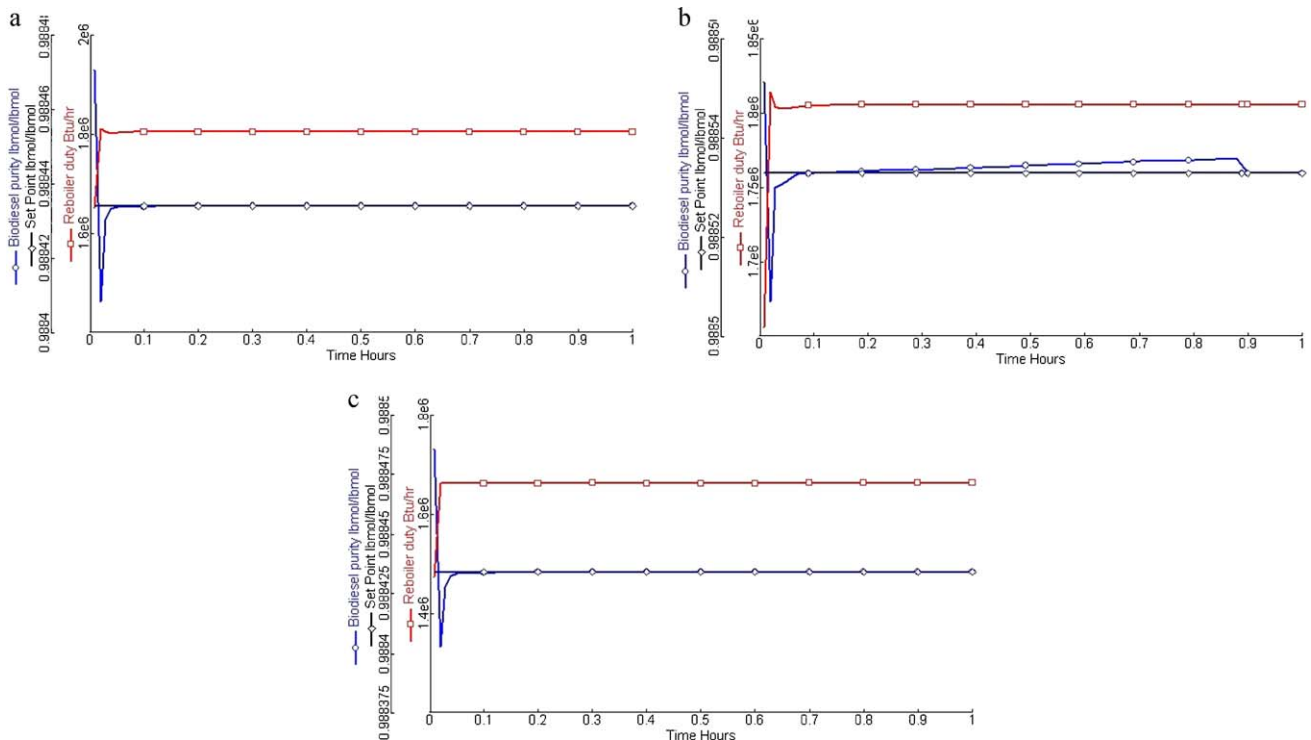


Fig. 9 – Closed loop dynamic response in STAIR for biodiesel stream in the design with (a) best SVD results, (b) minimum Q and (c) minimum IAE.

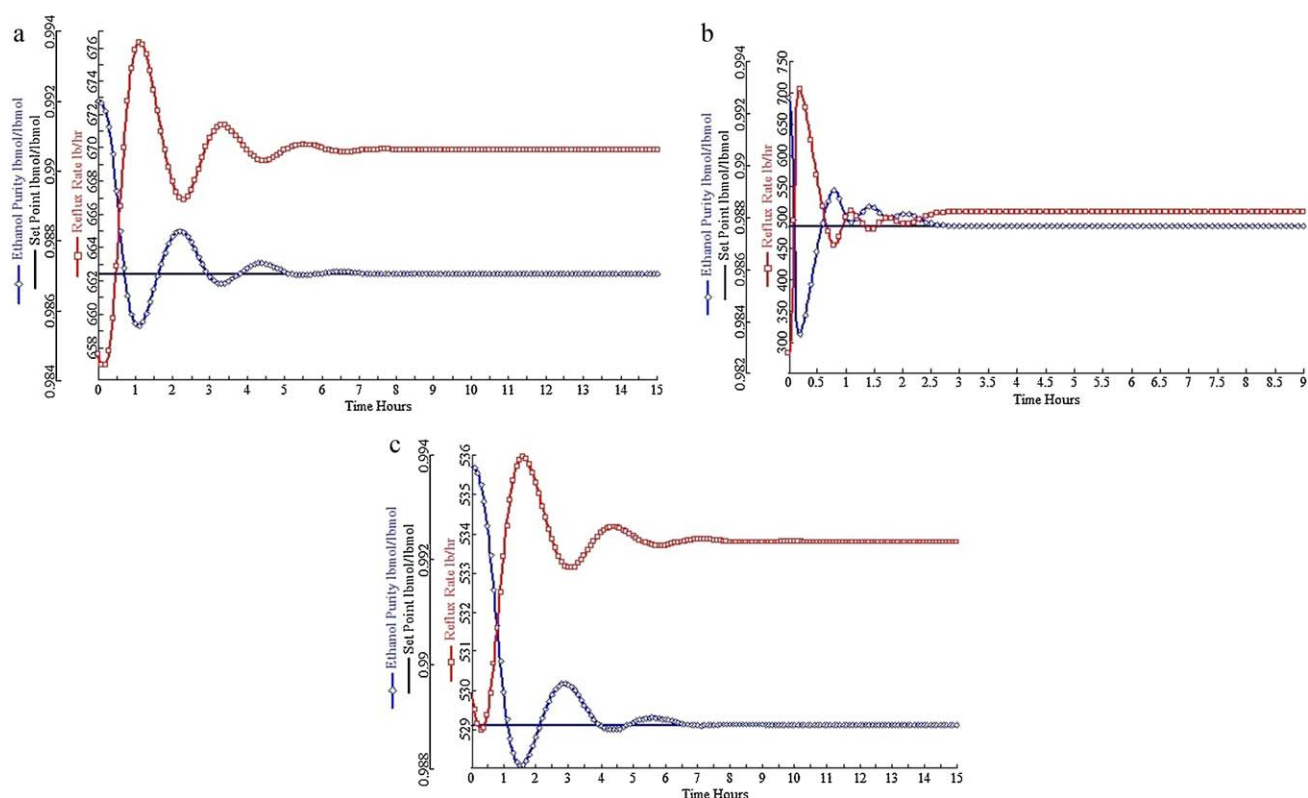


Fig. 10 – Closed loop dynamic response in SDCE for bioethanol stream in the design with (a) best SVD results, (b) minimum Q and (c) minimum IAE.

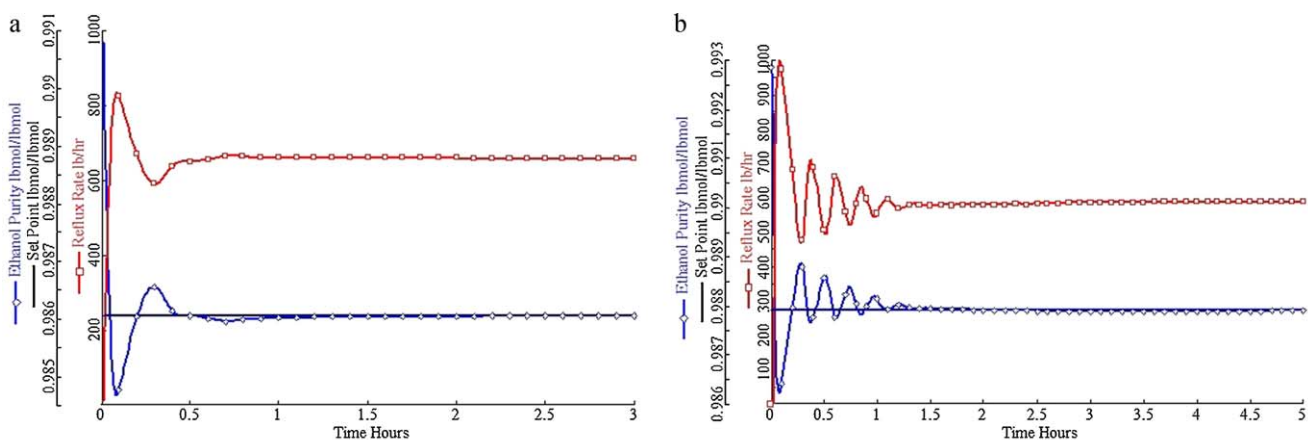


Fig. 11 – Closed loop dynamic response in STADE for bioethanol stream in the design with (a) best SVD results and minimum IAE (b) minimum Q.

designs. The arrangements with minimum energy consumption present worse control properties in comparison with schemes with high reboiler duty.

6. Conclusions

Upon analysis of the SVD and dynamic simulations, the controllability of coupled reactive and extractive schemes in different design conditions are compared for a given separation problem. At minimum energy consumption, number of stages and total annual cost conditions, the controllability is worse than the controllability in high energy consumption conditions. Therefore, the arrangements operating at “non-optimal conditions”, their controllability is better. The results obtained using SVD are similar to the results obtained using rigorous dynamic simulations. In general, the result is very

important because it indicates that intensified systems with side columns operated at high energy consumption conditions present the best controllability in comparison with designs with minimum energy consumption, total annual cost and number of stages.

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