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STUDY OF CONTROL PROPERTIES OF INTENSIFIED THERMALLY COUPLED DISTILLATION SEQUENCES

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Abstract
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. All sequences have been designed using a multi objective 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. 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 nonoptimal design conditions.

Keywords: process intensification, thermally coupling, 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[1]. Reactive and extractive distillations are classical examples of PI. 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; (3) (2) Direct heat integration decreases the heat demand. Motivated by the large energy requirements of distillation, researchers have developed several column arrangements that can bring savings in both energy and capital cost. Any reduction in energy consumption will not only bring economic benefits but also environmental benefits in terms of reduction in fossil fuel usage and their associated emissions. Reported studies reveal that the thermally coupled distillation systems (TCDS) provide the energy reduction in distillation columns[2]. TCDS 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. 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 savings that can be translated into reductions of total annual cost and miniaturization of the schemes. Those complex configurations offer an alternative to conventional reactive and extractive distillation towers or multicolumn arrangements, with potential for significant cost savings, according to the principles of PI.

Design issues for reactive and extractive distillation systems are significantly more complex than those involved in ordinary distillation. The optimization of a complex distillation system is usually characterized as being of large problem size, since the significant number of strongly nonlinear

equations results in serious difficulty in solving the model. Several optimization methods that utilize mathematical programming models for the design of reactive and extractive distillation columns have been proposed[3,4]. 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. In addition to the time and expertise needed to formulate these models, the synthesis and design of distillation sequences pose other difficulties, such as the presence of two or more (often conflicting) objectives to be simultaneously optimized. Finally, additional convergence problems are generated when discontinuous functions are introduced in the model. To compensate for these difficulties, it is often necessary to supply initial values for the optimization variables very close to the actual solution, something that is not always an easy task. In general, the optimal design of reactive or extractive complex distillation systems is a highly non-linear and multivariable problem, with the presence of both continuous and discontinuous design variables; also, the objective function used as optimization criterion is generally non-convex with several local optimums and subject to several constraints. 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 (black-box model). First, due to the fact that they are based on a direct search method, it is not necessary to have explicit information on the mathematical model or its derivatives. Secondly, the search for the optimal solution is not limited to one point but rather relies on several points simultaneously; therefore the knowledge of initial feasible points is not required and such points do not influence the final solution.

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: the extractive distillation of a mixture of ethanol-water using as entrainer ethylene glycol (Figure 1) and the production of biodiesel using reactive distillation (Figure 2) in comparison with conventional configurations. All sequences have been designed using a multi objective 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.

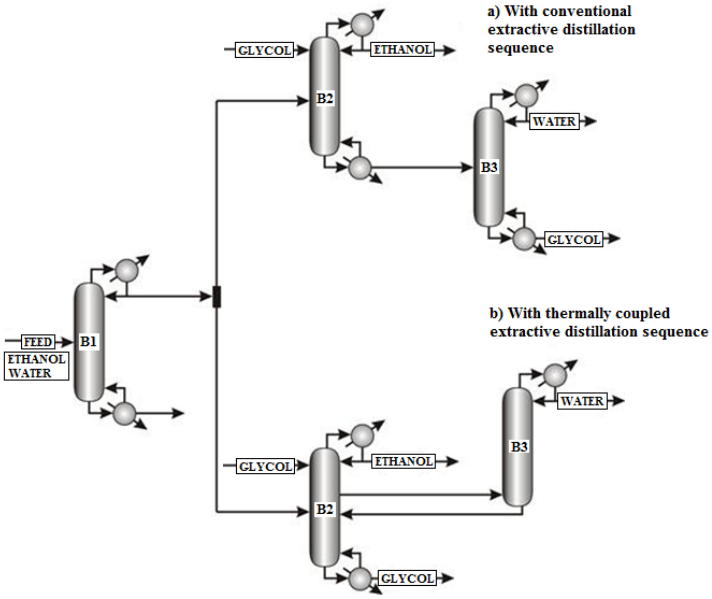


Figure 1. Process for the purification of bioethanol using schemes with: a) conventional extractive distillation structure and b) thermally coupled extractive distillation sequence.

2. Optimization Strategy

For the conventional and complex 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:

$$\text{Min}\,(Q_i, N_i, F_{EA}) = f(R_i, N_i, N_{F,i}, F_{EA}, F_k, N_k, F_{PS,o})$$

subject to

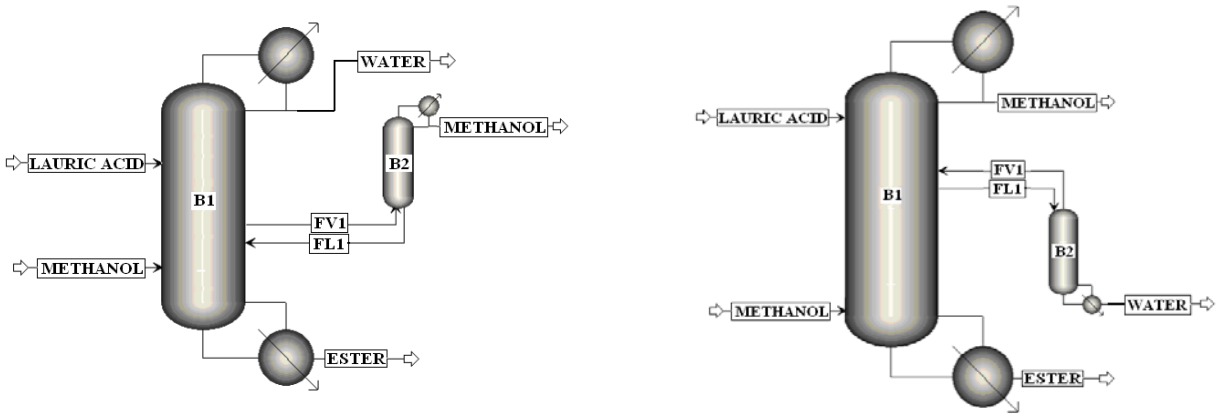
$$\vec{y}_m \geq \vec{x}_m$$

or

$$\text{Min}\,(Q_1, Q_2, N_1, N_2, N_R) = f(R_i, N_i, N_{F,i}, N_{r1}, N_{r2}, F_{PS})$$

subject to

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



a) With side rectifier (SDCR) b) With side stripper (STADR)

Figure 2. Process for the production of biodiesel using
a) conventional schemes and b) configuration with thermally coupling with a side column.

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, 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 four objectives to minimize: the number of stages, the extracting agent flow or reactive stages, and the heat duty of the sequence. For these sequences the objectives are in competition, so they have to be optimized simultaneously. The manipulated 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 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; from this initial solution 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. For more detailed information about this algorithm and its link to Aspen Plus, the reader is referred to the original work[5].

3. Control Analysis

Open loop dynamic responses to changes around the assumed operating point (for each configuration) were obtained. 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):

$$G = V\Sigma W^H \qquad (1)$$

where Σ = diag (σ_1,.....,σ_n), σ_ι = singular value of G = $\lambda_i^{1/2}$ (GG^H); V = (v_1, v_2,…..) matrix of left singular vectors, and W = (w_1, w_2,….) 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:

$$\gamma = \sigma^* / \sigma_* \quad (2)$$

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

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 proportional-integral 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 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 atm, and the second is methanol with a flow of 54.48 kmol/h as saturated vapor at 1.5 atm. 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[6] was used. 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 45.4 kmol/h ethanol in water [10% in moles of ethanol in water. This is a typical yield for fermentation of sugarcane bagasse] 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 (Figure 1a) 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 (Figure 1b), in the extractive stage of the separation, use a thermally coupled extractive distillation scheme. 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. Figure 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. Similarly, in the case of extractive configurations, the Pareto front shows that the configuration with thermally coupled extractive sequence provides energy savings of ~30% with respect to the best energy – efficient sequence based on conventional extractive distillation columns. These savings are not obtained by an increase in the total number of stages of the scheme. The theoretical control properties of thermally coupled distillation sequences were obtained using SVD technique. 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 number). In the Figure 4, the σ_* and γ for all cases of study are showed. There are important differences between the design operated at minimum energy consumption and the scheme operated at high total annual cost.

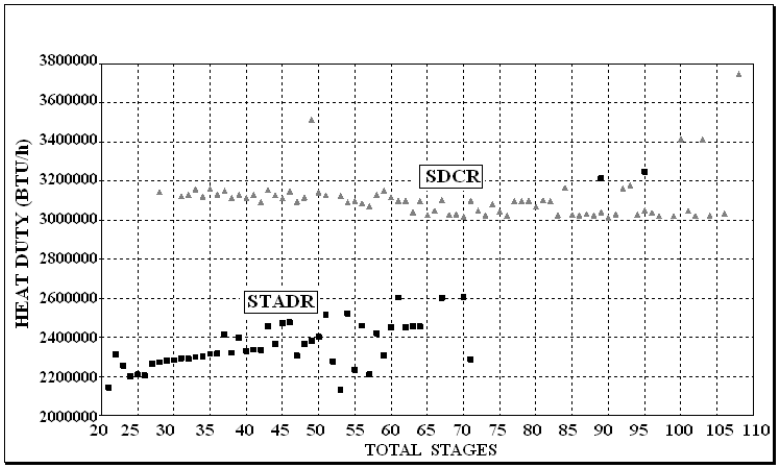


Figure 3. Total number of stages versus energy consumption of thermally coupled (STADR) and conventional reactive direct sequences (SDCR).

In the case of STADR and STAIR, when they are operated at nonoptimal conditions (high energy consumption, number of stages and total annual cost) their controllability improves (for example, design STAIR 16 is a nonoptimal design and the scheme STAIR 18 shows the minimum energy consumption for this scheme). In those nonoptimal conditions, STADR and STAIR present highest values of the minimum singular value (Figure 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 feedback control. The results for the condition number show that coupled sequences in the nonoptimal value offer the best value (Figure 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 optimal arrangement (in other words, the complex system can eliminate the disturbances better that the conventional arrangements in a non optimal operating 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 and number of stages and number of reactive stages are increased, the controllability improves. 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. 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 presents highest value and the condition number shows the minimum value. Also, the reboiler duty in the coupled systems is lower than the conventional sequence in the non optimal case.

The closed loop analysis was based on proportional-integral (PI) controllers. 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 the closed-loop analysis, several issues must be defined first, such as the control loops for each system, the type of process controller to be used, and the values of the controller parameters. Several techniques, such the relative gain array method, can be used to fix the loops for a control system. In the case of distillation columns, however, such loops are fairly well established and used successfully in industrial 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. The control loops for the systems, in a first option, 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 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. Figure 6 displays the IAE value for biodiesel stream. The results show that the scheme in nonoptimal

conditions offers the best dynamic behavior (minimum IAE value). This situation corroborates that operating in nonoptimal conditions is a good option. Similar results were obtained in the STADR scheme and coupled extractive configurations.

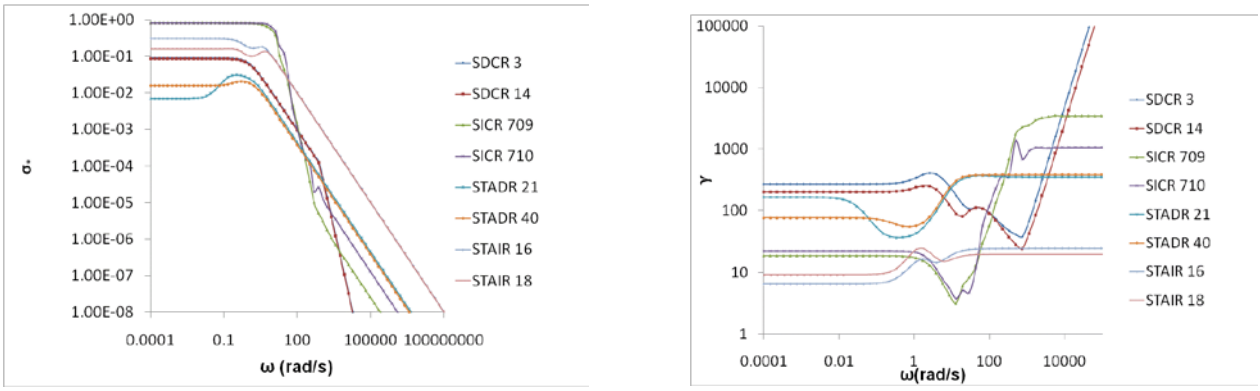
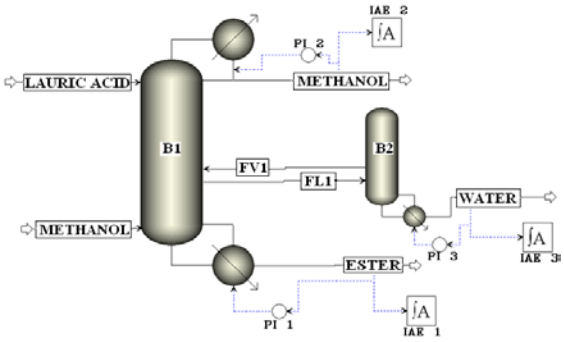


Figure 4. Minimum singular value and condition number for different thermally coupled reactive schemes with side rectifier (STADR), side stripper (STAIR), conventional reactive direct sequence (SDCR) and indirect sequence (SICR).



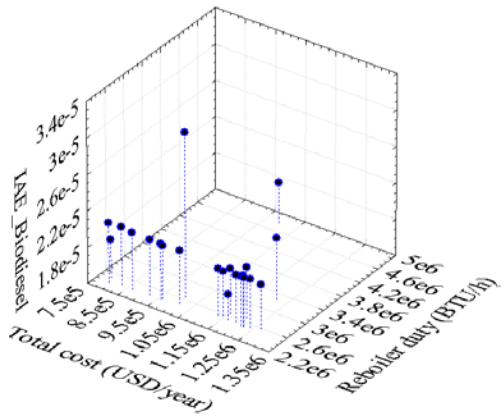


Figure 5. Control loops proposed for STADR. **Figure 6.** IAE values for biodiesel component in STAIR.

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 optimal design (minimum energy consumption, number of stages and total annual cost); the controllability is worse than the controllability in nonoptimal conditions. The closed-loop dynamic simulations corroborate the theoretical control properties obtained using the singular value decomposition technique. In general, the results are important because indicate that intensified systems with side columns operated at nonoptimal design conditions present the best controllability in comparison with designs with minimum energy consumption, total annual cost and number of stages.

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