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Controllability analysis of thermodynamically equivalent thermally coupled arrangements for quaternary distillations

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ABSTRACT

It has been proved that thermally coupled configurations can lower the energy consumptions around 30% in comparison to conventional distillation schemes for the separation of ternary mixtures. These potential energy savings have motivated research efforts on thermally coupled configurations for four or more component mixtures. Besides the significant energy savings that they have in contrast to conventional distillation sequences, one distinct feature of a thermally coupled configuration is that there is the flexibility to rearrange the column sections among the thermodynamically equivalent structures. A thermodynamically equivalent structure is a distinct conceptual design in terms of the column equipment and their interconnections, which provide the opportunity for optimal equipment design, as well as for the improvement of the dynamics and operability of the thermally coupled configurations.

In this paper, a detail dynamic analysis of the thermodynamically equivalent structures for four-component mixtures is presented. The simulation results for the various cases of study have shown that the thermodynamically equivalent structures have different dynamic closed-loop responses in contrast to the corresponding original thermally coupled configurations. It has been observed that the dynamic responses depend on the thermodynamically equivalents structures and their corresponding counterparts of the original thermally coupled configurations. Therefore, for a thermally coupled configuration with substantial energy savings, it is significant to compare the dynamic responses of the thermodynamically equivalent structures to find the optimal solution in terms of both energy consumption and closed-loop dynamic performance.

1. Introduction

Thermally coupled configurations (TCDS) can improve the intrinsic separation efficiency of the traditional distillation configurations due to the avoiding of the remixing (Triantafyllou and Smith, 1992). Therefore, they have the advantages to save both energy and capital costs in comparison to the traditional distillation sequences (Tedder and Rudd, 1978; Smith and Linnhoff, 1988; Flores et al., 2003). For example, for ternary distillations, the thermally coupled configurations are capable of achieving energy savings of up to 30% compared to the conventional direct and indirect distillation sequences. Because there are only a few possible alternatives of thermally coupled configurations for ternary mixtures, a large number of works have been done in the analysis of both steady-state and dynamic performance. From which, the dividing wall column (DWC) has been successfully used in many industrial applications, where 30–50% of both energy and capital cost savings have been achieved (Schultz et al., 2006; Kaibel et al., 2006). Recently, Malinen and Tanskanen (2007) have presented an optimization procedure to mimic Petlyuk columns with a

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Nomenclature

b	exergy function (Btu/h)
G	transfer function matrix
h	molar enthalpy (Btu/lb-mol)
K_C	proportional gain
LF	interconnecting liquid flow (kmol/h)
n	mole flow (lb-mol/h)
Q	Reboiler heat duty (Btu/h)
S	molar entropy (Btu/lb-mol R)
TAC	Total annual cost ($/year)
TCDS	thermally coupled distillation sequences
T_0	temperature of the surroundings (R)
T_s	temperature of the system (R)
VF	interconnecting vapor flow (kmol/h)
W_{min}	minimum work for the separation (Btu/h)
W_s	shaft work (Btu/h)

Greek symbols

γ^*	condition number
η	second law efficiency
σ^*	maximum singular value
σ_*	minimum singular value
τ_I	reset time (min)

high number of stages to separate zeotropic and azeotropic mixtures.

The successful application of the thermally coupled systems for ternary separations has motivated more efforts to study thermally coupled configurations for four or more components mixtures (Agrawal, 1996; Christiansen et al., 1997; Blancarte-Palacios et al., 2003; Rong and Kraslawski, 2003; Rong et al., 2003). However, it must be indicated that the recent works on thermally coupled configurations for four or more component mixtures are primarily focused on either the conceptual synthesis of the new structures or the preliminary screening among the alternatives for the economic evaluation based on shortcut models. These studies are mainly concerned with the steady-state design and energy performance. Nevertheless, the available works have contributed a significant progress in the synthesis and design of the thermally coupled configurations for four or more component mixtures.

For four or more component mixtures, a distinct feature of a thermally coupled configuration is that it can produce a quite large number of thermodynamically equivalent structures (TES). The TESs are produced by rearrangements of the column sections of the original thermally coupled configurations (OTC). Although there is the same nominal energy consumption between the TESs and the OTC, each feasible TES represents *a distinct new conceptual design* for the distillation system in terms of the actual columns and their interconnections. Therefore, it is paramount importance to study the TESs in order to find an optimal system for specific applications. For example, recent works have made significant progress in the synthesis of the thermodynamically equivalent structures for thermally coupled configurations for four or more component mixtures (Agrawal, 2000; Caballero and Grossmann, 2003; Rong and Turunen, 2006). During the synthesis of the feasible structures, some preliminary observations have been made for the TESs. Jiménez et al. (2003), Caballero and Grossmann (2003) have shown that there are more operable configurations among the TESs in terms of vapor transfers between the columns. Rong and Kraslawski (2003) have shown that the TESs can reduce the capital costs and improve column equipment design for hydraulic performance. These preliminary works have shown that the TESs have significant impact on both systems' design and operation. However, for specific applications, even though the steady-state design and preliminary screening can confirm the significant energy savings, it is useful and necessary to have the detail control properties comparison of the TESs and the OTC. This will guarantee the final optimal solution in terms of both steady-state and dynamic performance. Such kind of detail dynamic analysis is invariably implemented for the specific cases in order to do the detail comparison between the studied configurations. It is really important to determine the optimal distillation sequence in terms of both steady-state performance, i.e., minimum energy consumption and the theoretical properties and closed loop dynamic responses. It is clear that the dynamic responses depend on the design structure, therefore the formulation of the design problem must take into account both issues but the solution is really complicated. As a result, the solution can be conducted in two stages: in the first stage, the optimal design is obtained in terms of energy consumption and in the second stage; the theoretical control properties and dynamic closed loop responses are obtained. Finally, the distillation sequence with the minimum energy consumption and good dynamic properties is considered as the optimal solution.

In this work, the cases of study for the control properties analysis between TESs and OTC for quaternary distillations are implemented. The OTC and the TESs are generated for the conventional sequence of AB/CD → A/B → C/D (CS; Fig. 1): (a)

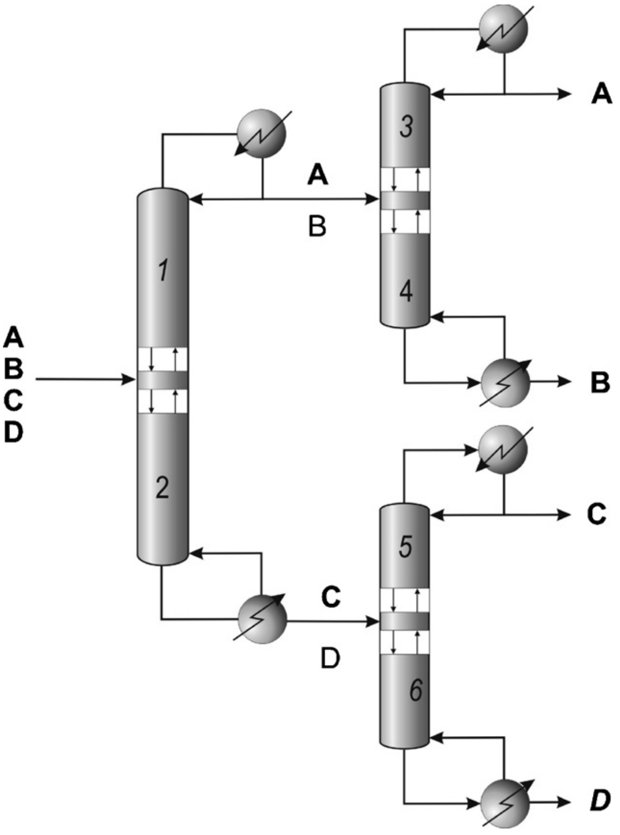


Fig. 1 – Conventional distillation sequence with prefactionator (CS).

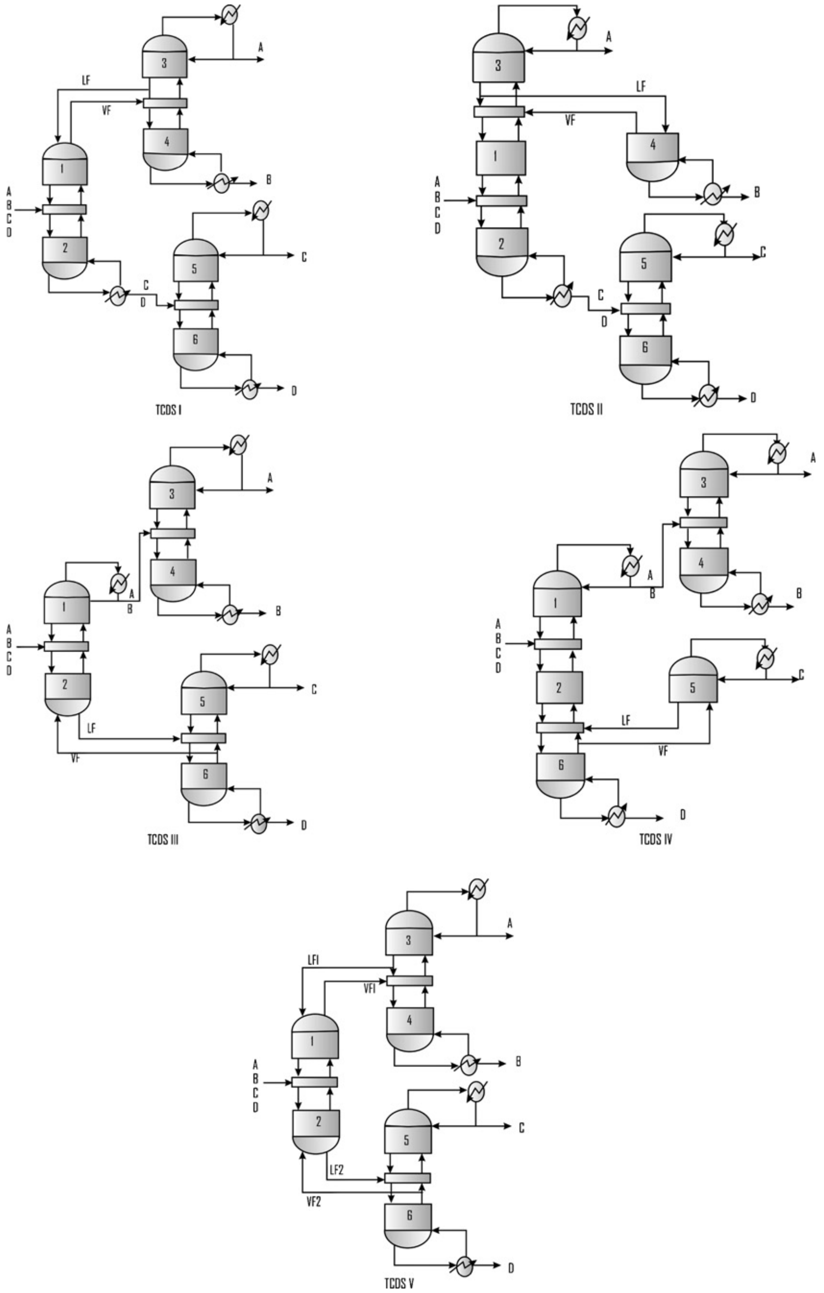


Fig. 2 – Thermally coupled distillation sequences for the separation of quaternary mixtures.

TCDS-I: just eliminate condenser in section 1 (Fig. 1) and introduce one thermal coupling (Fig. 2a); (b) TCDS-II: move column section 3 to the top of column, section 1, in CS (Fig. 2b); (c) TCDS-III: just eliminate reboiler in section 2 (Fig. 1) and introduce one thermal coupling CD (Fig. 2c); (d) TCDS – IV: move column section 6 to the bottom of column, section 2, in CS (Fig. 2d); (e) TCDS – V: eliminate both condenser and reboiler in section 1 and 2 of CS and introduce both thermal coupling (Fig. 2e). In these configurations, it is apparent that the TESs have different columns as well as different inter-column connections than the OTCs. It is also clear that in the TESs the products are produced from different columns than the OTCs. These differences will affect both steady-state design and dynamic properties of the thermally coupled configurations. For these configurations, we developed a comparative study of the energy consumption, thermodynamic efficiency, total annual costs and control properties (using the singular value decomposition technique in the frequency domain and rigorous closed-loop dynamic simulations). The cases are studied for different quaternary mixtures with different feed compositions. The results show that the structure has different effect on energy consumption, capital costs and dynamic performance.

2. Design and dynamic analysis of complex sequences

In this case, to overcome the complexity of the simultaneous solution of the tray arrangement and energy consumption within a formal optimization algorithm, we decoupled the design problem in two stages: (1) tray configuration; (2) energy-efficient design (optimal energy consumption). The first step for this analysis consists in the detection of a base design for

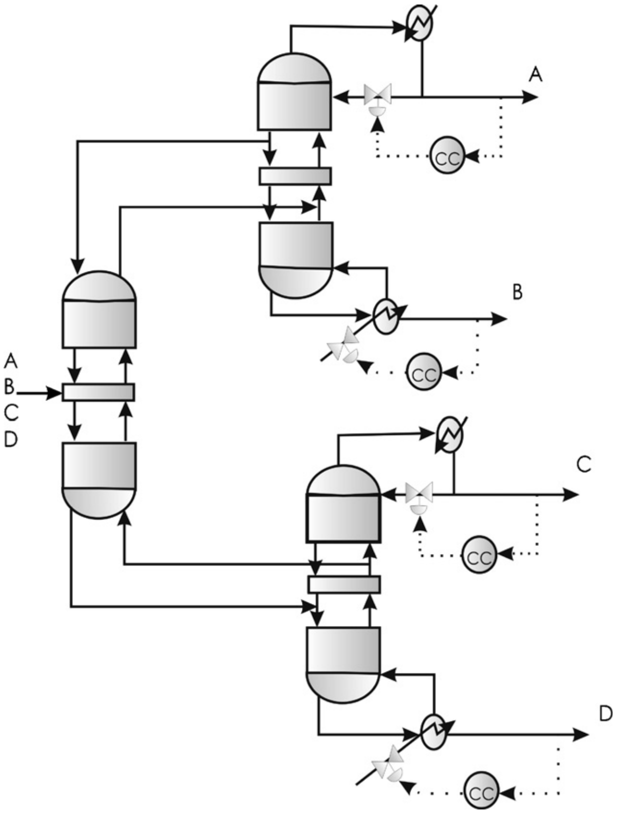
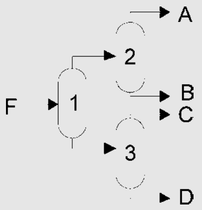


Fig. 3 – Control loops based on practical considerations for the TCDS-V scheme.

Table 1 – Design variables for the CS, mixture 1, Feed F1

	Column 1	Column 2	Column 3
Number of stages	22	18	17
Feed stage	12	10	8
Distillate rate (lbmol/h)	50.00	40.35	9.61
Bottom rate (lbmol/h)	50.00	9.65	40.39
Feed flow rate (lbmol/h)	100.00	50.00	50.00
Reflux ratio	1.70	0.96	34.59
Temperature of distillate (°F)	133.04	120.4	269.34
Pressure of top (psi)	68.74	66.24	71.24
Diameter (ft)	2.10	1.30	3.27

the integrated arrangements. The design of such schemes was carried out through a section analogy procedure, taking as a basis the tray structure of the conventional sequence given in Fig. 1. The conventional system contains six sections; those tray sections can also be identified within the structure of the five complex sequences (Fig. 2), thus providing the basis for the tray arrangement of the interconnected structure. Using the well-known short cut method of Fenske, Underwood and Gilliland (Seader and Henley, 2006); the tray structure of conventional distillations schemes was obtained. The number of the trays was obtained using a reflux ratio of 1.33 times the minimum value for each separation. To avoid the use of refrigerants that would have a high impact on the economics of the separation sequence, the design pressure for each column was chosen such that all condensers could be operated with cooling water. After the base designs for the TCDS structures were obtained, the degrees of freedom that remain are used to get the integrated designs that provide minimum energy consumption. One or two degrees of freedom (depending of the scheme, see Fig. 2) remain for each integrated sequence. They are the interconnecting flows: vapor (VF) or liquid (LF), depending of the scheme. The search procedure provides the optimal values of the interconnecting flows to minimize the energy consumption for the separation. The design is successful if it meets the product specifications. Further details on the design and optimization method are available in Blancarte-Palacios et al. (2003). We complement the study for the conventional and complex schemes calculating the total annual cost (TAC), according to Turton et al. (2004), and second law efficiency (η), following the method proposed by Seader and Henley (2006). More details are included in Appendices A and B.

The understanding of the dynamic behavior of distillation columns has received considerable attention due to the fact that distillation is one of the most widely used unit operations in the chemical process industries. As the disturbances in process variables under actual operation conditions are almost inevitable, the prediction of the transient responses of a distillation column bears much importance in the sense of the effective control of the separation process (Berber and Karadurmus, 1989). Such resulting structures with minimum energy consumption provided the designs that were subjected to the dynamic analysis. Open loop dynamic responses around the assumed operating point (which corresponds to that with minimum energy consumption for each configuration) were obtained. The responses were obtained through the use of Aspen Dynamics 11.1 by implementing step changes in the manipulated variables and registering the open loop dynamic response of each composition. Also, state space matrices can be obtained in a similar way. Transfer functions were grouped into transfer function matrices (G) and they were subjected to singular value decomposition (SVD) in the frequency domain (for more details about SVD see Klema and Laub, 1980). Two parameters of interest are the minimum singular value, σ_*, and the ratio maximum (σ^*) to minimum singular values, or condition number (γ^*). The condition number reflects the

Table 2 – Energy consumption, total annual cost and thermodynamic efficiency (case M1F1)

Arrangement	Energy consumption, (Btu/h)	TAC ($/year)	η (%)
CS	5,793,805.67	1,132,804	12.02
TCDS-I	2,410,588.21	279,829	20.98
TCDS-II	2,412,427.45	281,508	21.13
TCDS-III	3,698,480.47	956,179	19.08
TCDS-IV	3,645,163.12	990,869	19.67
TCDS-V	4,733,673.25	963,960	14.24

Table 3 – Energy consumption, total annual cost and thermodynamic efficiency (case M1F2)

Arrangement	Energy consumption, (Btu/h)	TAC ($/year)	η (%)
CS	6,353,675.67	930,937	10.23
TCDS-I	3,616,224.94	892,882	15.33
TCDS-II	3,618,082.31	856,721	15.45
TCDS-III	3,666,761.62	894,811	17.50
TCDS-IV	3,625,710.96	872,244	17.33
TCDS-V	2,980,589.18	791,242	20.46

Table 4 – Energy consumption, total annual cost and thermodynamic efficiency (case M2F1)

Arrangement	Energy consumption, (Btu/h)	TAC ($/year)	η (%)
CS	8,777,198.10	1,852,833	6.41
TCDS-I	5,993,797.86	1,814,209	6.91
TCDS-II	5,893,473.45	1,804,429	6.89
TCDS-III	7,480,929.11	1,772,593	7.44
TCDS-IV	7,496,773.61	1,750,163	7.45
TCDS-V	6,663,682.92	1,741,541	8.06

Table 5 – Energy consumption, total annual cost and thermodynamic efficiency (case M2F2)

Arrangement	Energy consumption, (Btu/h)	TAC ($/year)	η (%)
CS	14,756,133.68	2,723,117	3.81
TCDS-I	9,467,232.64	1,755,075	4.82
TCDS-II	9,575,794.45	1,807,865	4.44
TCDS-III	9,683,117.75	2,378,612	4.75
TCDS-IV	9,838,675.70	2,338,881	4.46
TCDS-V	9,783,404.29	2,398,367	4.69

Table 6 – Energy consumption, total annual cost and thermodynamic efficiency (case M3F1)

Arrangement	Energy consumption, (Btu/h)	TAC ($/year)	η (%)
CS	6,809,276.99	2,207,084	12.94
TCDS-I	5,933,797.86	2,160,530	14.46
TCDS-II	5,820,845.73	2,112,621	14.50
TCDS-III	6,218,114.84	1,871,747	17.47
TCDS-IV	N/A	N/A	N/A
TCDS-V	N/A	N/A	N/A

N/A: not available.

sensitivity of the system under uncertainties in process parameters and modeling errors. 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. 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.

The study was complemented with a closed-loop dynamic analysis. This analysis was based on proportional-integral (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

Table 7 – Energy consumption, total annual cost and thermodynamic efficiency (case M3F2)

Arrangement	Energy consumption, (Btu/h)	TAC ($/year)	η (%)
CS	9,827,717.94	2,387,609	7.41
TCDS-I	6,851,350.49	1,993,083	11.91
TCDS-II	6,843,669.77	2,074,983	11.89
TCDS-III	7,329,498.28	1,982,781	13.34
TCDS-IV	N/A	N/A	N/A
TCDS-V	7,109,670.10	1,836,361	13.27

N/A: not available.

Table 8 – Transfer function matrix for CS (M1, F1)

$$\begin{array}{c} \\ A \\ B \\ C \\ D \end{array} \begin{array}{c} \begin{array}{cccc} R1 & Q1 & R2 & Q2 \end{array} \\ \begin{pmatrix} \frac{-0.0048}{1+68.009766s} & \frac{-2.1252}{1+0.34683s} & \frac{-0.0028}{1+1.963915s} & \frac{0.0048e^{-1.08s}}{1+1.62s} \\ \frac{0.0176}{1+2.382s}-\frac{0.0064}{1+6.21857s} & \frac{2.6136}{(1+0.22089s)^2} & \frac{0.0164}{1+2.31s}-\frac{0.0064}{1+6.21s} & \frac{0.0172}{1+2.16253s}-\frac{0.0068}{1+5.8374s} \\ \frac{0.0032e^{-1.16s}}{1+0.58s} & \frac{0.0036e^{-1.16s}}{1+0.867044s} & \frac{0.0036e^{-1.08s}}{1+1.0502s} & \frac{-10.1712}{(1+0.67667s)^2} \\ \frac{-0.0068}{1+4.64s}+\frac{0.0056}{1+18.25s} & \frac{-0.006}{1+4.866s}+\frac{0.004}{1+17.06923s} & \frac{-0.006}{1+4.82s}+\frac{0.004}{1+16.8291s} & \frac{3.188}{(1+0.42453s)^2} \end{pmatrix} \end{array}$$

multivariable control problem. For these cases, the tuning procedure was conducted taking into account one control loop at a time; the parameters thus obtained were taken for the following control loop until the four loops were closed. For the dynamic analysis, individual set point changes for product composition were implemented for each of the four product streams. One of the key parts for the dynamic analysis is the selection of control outputs and manipulated variables for each control loop. We based our selection in practical consideration. The pairings in the control loops for the distillation columns were the corresponding reflux flowrates to control the compositions A and C, while the reboiler heat duties were used for the control of B and D. At first instance, standard control loops (top product-reflux flowrate and bottom product-reboiler heat duty) were used for each column (See for example Fig. 3). Second, we obtained the pairing of same manipulated and control variables using the relative gain array method (RGA).

3. Cases of study

The cases of study were selected to reflect different separation difficulties and different contents of the intermediate component of the quaternary mixtures. Three mixtures were considered: *n*-butane, *n*-pentane, *n*-hexane and *n*-heptane (M1); benzene, toluene, ethyl-benzene and *o*-xylene (M2); *i*-butane, *n*-butane, *i*-pentane and *n*-pentane (M3), with a feed

Table 9 – Transfer function matrix for TCDS - I (M1, F1)

$$\begin{array}{c} \\ A \\ B \\ C \\ D \end{array} \begin{array}{c} \begin{array}{cccc} R1 & Q1 & R2 & R3 \end{array} \\ \begin{pmatrix} \frac{-0.0172}{1+0.712s} & \frac{-0.8396}{1+0.801s} & \frac{-0.0172}{1+0.731s} & \frac{-0.0172}{1+0.88644s} \\ \frac{-0.0556}{1+9.009629s} & \frac{0.3576}{1+1.29827s}-\frac{1.4612}{(1+3.3734s)^2} & \frac{-0.0552}{(1+4.8270635s)^2} & \frac{-0.0548}{1+8.9802424s} \\ \frac{-0.0084}{1+11.79s} & \frac{-0.3072}{(1+1.791s)^2} & \frac{-0.0084}{1+13.1s} & \frac{-10.2716}{1+1.30476s} \\ \frac{0.0252e^{-2.84s}}{1+8.46s} & \frac{0.4556}{(1+1.98801s)^2} & \frac{0.0256e^{-2.6s}}{1+8.700972s} & \frac{3.2408}{1+0.853983s} \end{pmatrix} \end{array}$$

Table 10 – Transfer function matrix for TCDS - III (M1, F1)

$$\begin{array}{c|cccc}
 & R1 & Q1 & R2 & Q2 \\
\hline
A & \dfrac{-0.0044}{1+0.974s}+\dfrac{0.0012}{1+2.2736s} & \dfrac{-2.1252}{1+0.636s} & \dfrac{-0.004}{1+5.312s}+\dfrac{0.0008}{1+11.216827s} & \dfrac{0.7164}{1+0.897s}-\dfrac{0.1064}{1+1.73s} \\
B & \dfrac{0.0316}{1+5.03s}-\dfrac{0.0108}{1+12.757543s} & \dfrac{2.4688}{1+0.444s} & \dfrac{0.0296}{1+5.897575s}-\dfrac{0.0096}{1+14.79875s} & \dfrac{-18.2968}{1+2.03615s} \\
C & \dfrac{-0.0128}{1+5.8s}+\dfrac{0.002}{1+11.354213s} & \dfrac{-0.0128}{1+5.75s}+\dfrac{0.002}{1+11.354213s} & \dfrac{-0.0128}{1+5.6596s}+\dfrac{0.002}{1+11.170887s} & \dfrac{-9.5244}{1+1.50396s} \\
D & \dfrac{0.0088}{1+3.93s} & \dfrac{0.0088}{1+3.42s} & \dfrac{0.0088}{1+3.012s} & \dfrac{3.0528}{1+1.01841s}
\end{array}$$

flowrate of 100 lb mol/h. To examine the effect of the content of the intermediate components, two types of feed compositions (in mole fraction) were assumed F1 (0.40/0.10/0.10/0.40) and F2 (0.10/0.40/0.40/0.10). The specified product purities for components A, B, C and D were 98.7, 98, 98 and 98.6%, respectively.

4. Results

The resulting designs and their performance with respect to energy consumption, thermodynamic efficiency, TAC and dynamic behavior are discussed in the following sections.

4.1. *Energy consumption and economic analysis*

The first part of the analysis was conducted to detect the designs with minimum energy consumption for the integrated sequences. The results were obtained by using an optimization procedure to detect the optimum values of the recycle streams for each of the three thermally coupled distillation sequences. It is well-known that, in a thermally coupled configuration, one can generate thermodynamically equivalent structures by rearrangements of some of the column sections among the column units. For example, the generation of thermodynamically equivalent structures from the original thermally coupled configurations for

Table 11 – Transfer function matrix for TCDS - V (M1, F1)

$$\begin{array}{c|cccc}
 & R1 & Q1 & R2 & Q3 \\
\hline
A & \dfrac{-1.7752e^{-10.5s}}{(1+5.743066s)^2} & \dfrac{-3.7584}{1+3.87573s} & \dfrac{-1.7748e^{-11.3s}}{(1+1.29675s)^2} & \dfrac{-5.7508}{(1+0.4994s)^2}+\dfrac{0.9804}{1+1.62036s} \\
B & \dfrac{-12.0248e^{-12.68s}}{(1+5.147465s)^2} & \dfrac{-20.6076}{(1+2.5001s)^2} & \dfrac{-12.0228e^{-12.68s}}{(1+5.208445s)^2} & \dfrac{-33.1872}{(1+0.4994s)^2}+\dfrac{3.7012}{1+1.434s} \\
C & \dfrac{-0.4236e^{-15.04s}}{(1+5.211506s)^2} & \dfrac{-0.9188}{(1+3.00193s)^2} & \dfrac{-0.4212e^{-14.18s}}{(1+5.431005s)^2} & \dfrac{-17.7392}{(1+1.077208s)^2} \\
D & \dfrac{0.6516e^{-12.46s}}{(1+5.0231s)^2} & \dfrac{1.0716}{(1+2.337512s)^2} & \dfrac{0.6516e^{-11.76s}}{(1+5.3s)^2} & \dfrac{4.1116}{1+1.0632641s}
\end{array}$$

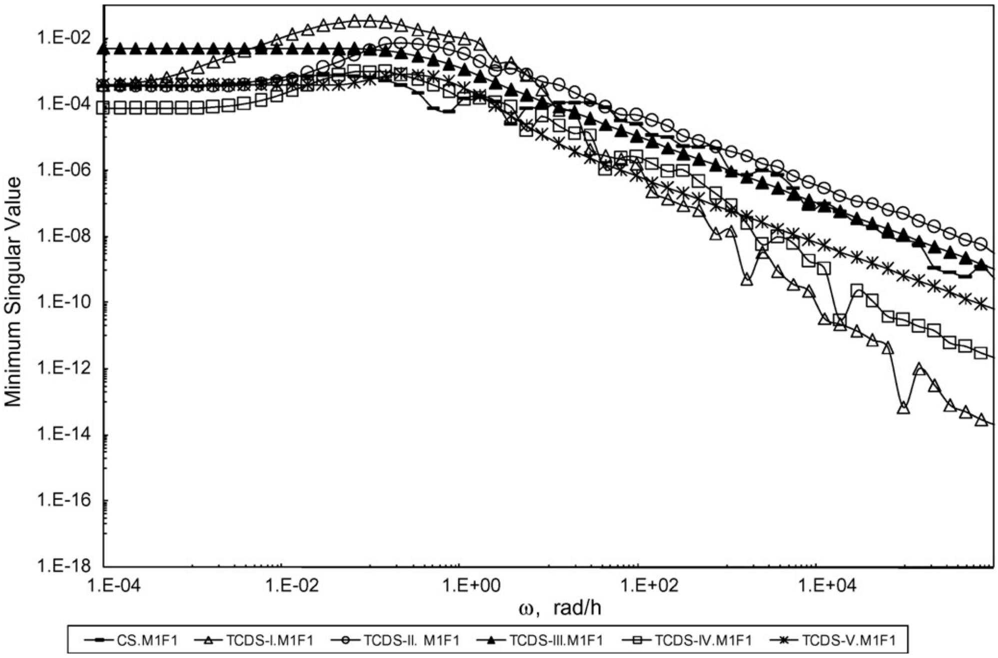


Fig. 4 – Minimum singular values (M1, F1).

ternary mixtures was illustrated by Rong and Kraslawski (2002). A thermodynamically equivalent structure is defined as a thermally coupled configuration that has the same individual splits and the same thermal couplings for the same submixtures, while it contains different structural arrangements of the column sections among the column units from the original thermally coupled configuration (Rong et al., 2003). It is possible to generate different thermodynamically equivalent structures for four or more component mixtures. For example, starting from the TCDS-I (Fig. 2a) we generated its thermodynamically equivalent structure TCDS-II (Fig. 2b). In the same form, starting from the TCDS-III (Fig. 2c), the thermodynamically equivalent structure TCDS-IV (Fig. 2d) can be obtained.

The tray arrangements and some important design variables for CS (case M1F1) are given in Table 1. The minimum energy requirements (details on the optimization procedure can be found in Blancarte-Palacios et al., 2003), TAC and η for each integrated scheme and conventional sequence for the three quaternary mixtures and the two assumed feed compositions are displayed in Tables 2–7. The results can be summarized as follow:

- The results indicate that the thermally coupled distillation sequences can offer energy savings between 10 and 40% in

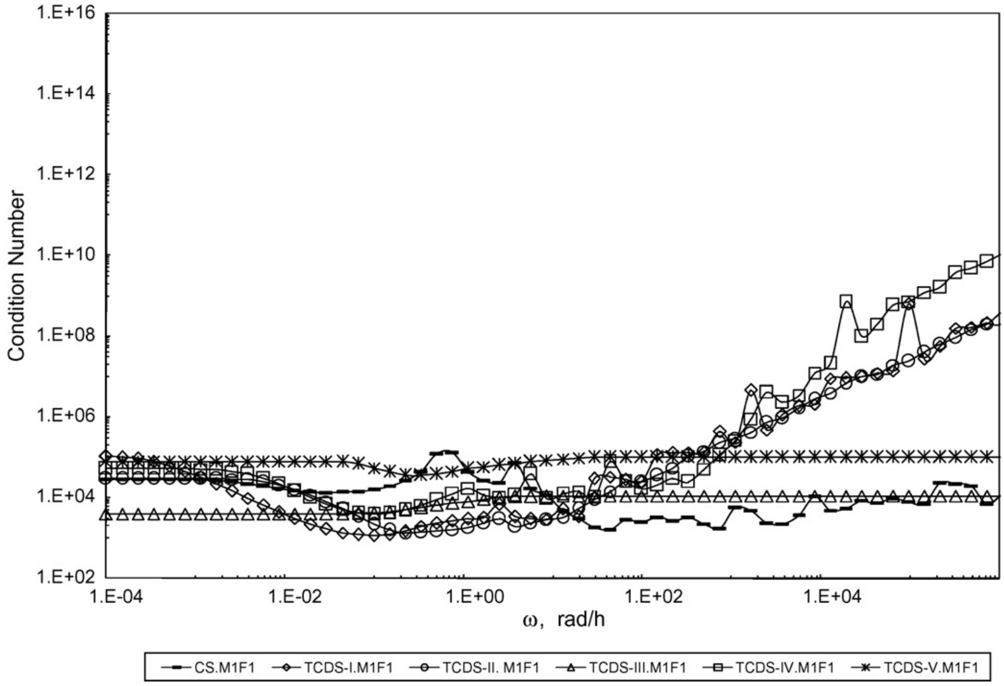


Fig. 5 – Condition numbers (M1, F1).

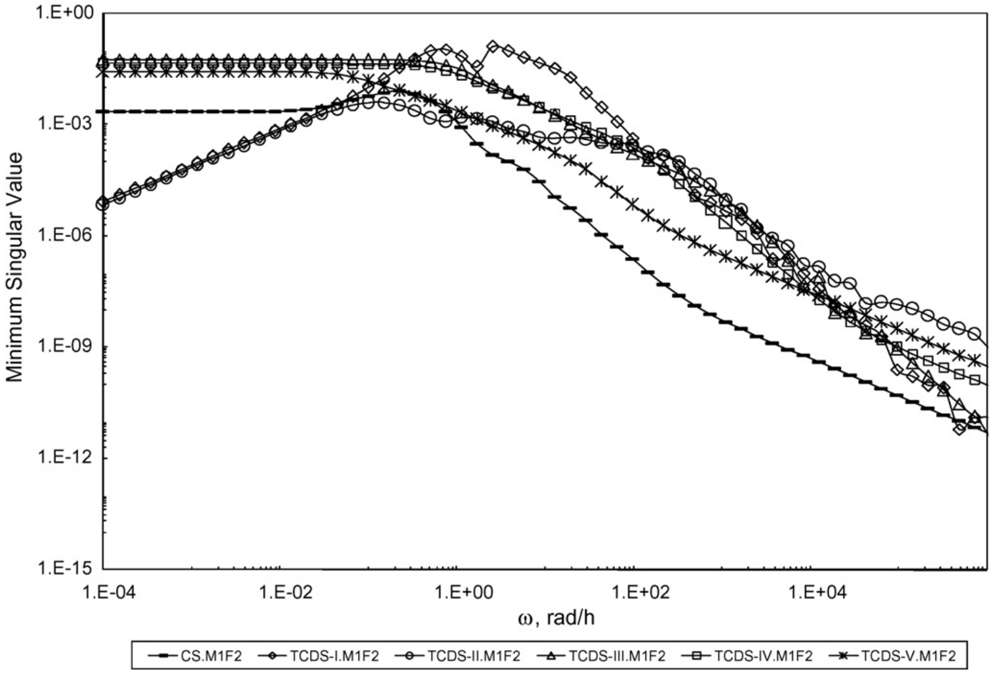


Fig. 6 – Minimum singular values (M1, F2).

contrast to the conventional distillation option (CS). This situation is function of the mixture and composition in the feed.

- The efficiency (η) in the use of the energy, in comparison with CS, is better or similar in the thermally coupled distillation sequences.
- TCDS-I and TCDS-II are thermodynamically equivalent structures. In all cases the results show similar energy consumption and η.
- TCDS-III and TCDS-IV are thermodynamically equivalent arrangements. The results displayed in Tables 2–7 show similar energy consumption and η.
- In all cases, the complex distillation sequences have the lowest values of TAC.

Based on the trends observed, some general aspects can be detected: (i) the use of thermal linking in distillation sequences offer energy savings, better or similar values of η and lower TAC values in comparison with CS; (ii) the lowest values of energy consumption can be obtained when component B is purified in a scheme with side stripper (TCDS-II) or its thermodynamically equivalent structure. In other words, the structure has different effect on energy consumption and η for a specific separation.

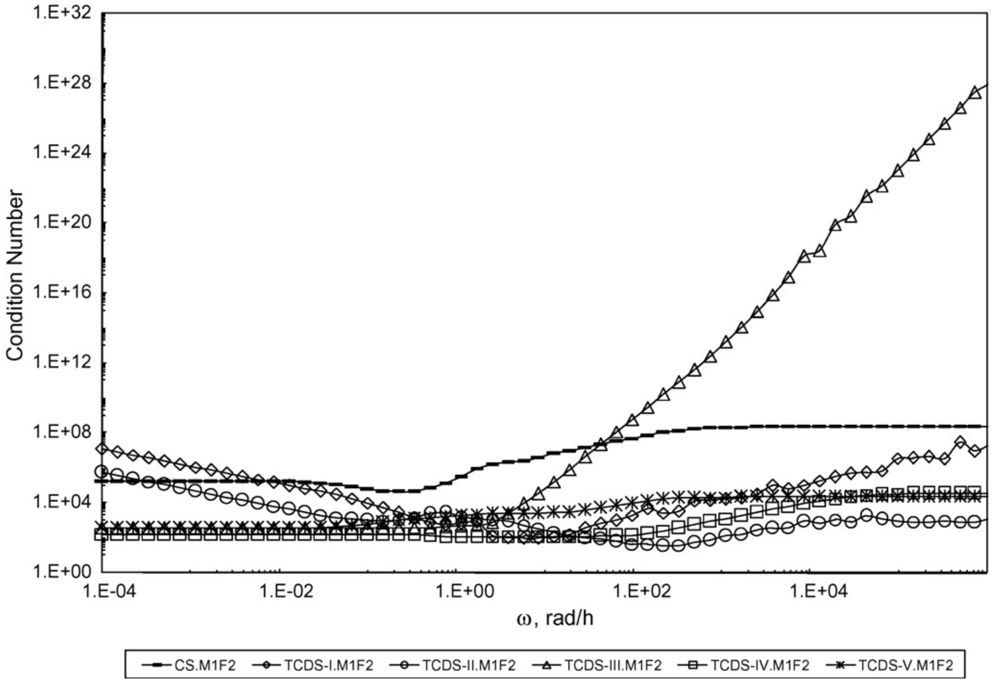


Fig. 7 – Condition numbers (M1, F2).

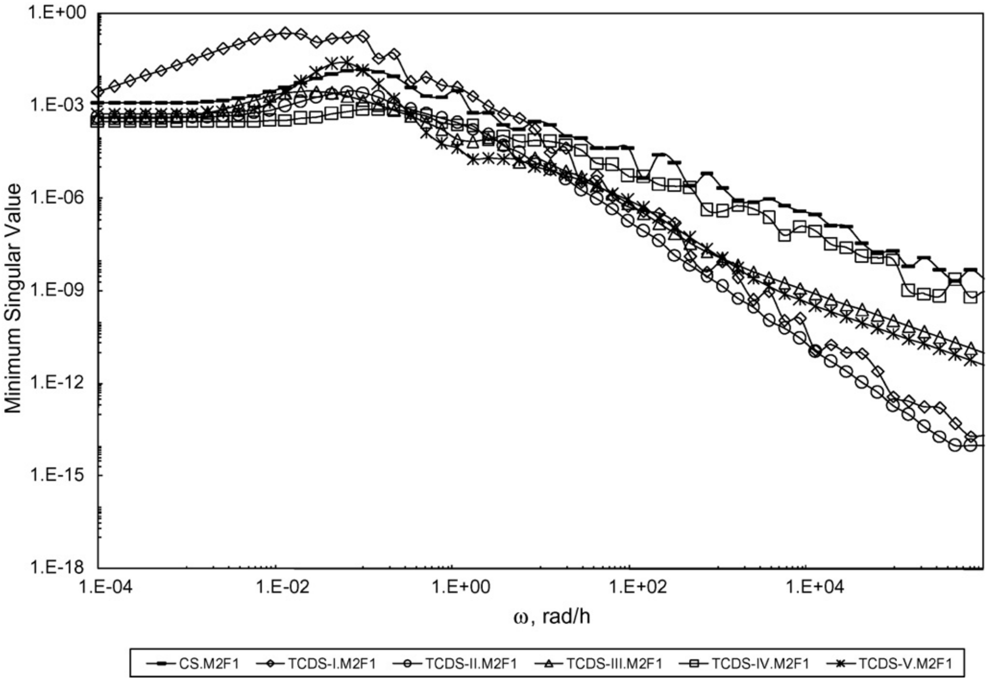


Fig. 8 – Minimum singular values (M2, F1).

4.2. SVD analysis

The SVD technique requires transfer function matrices, which are generated by implementing step changes in the manipulated variables of the optimum design of the distillation sequences and registering the dynamic responses of the four products. It can be mentioned that for a system with a single input and a single output, the zeros and poles of the transfer function give important information in order to evaluate the fully controllability of the system, but for multivariable systems, the SVD analysis provides the theoretical control properties. For the distillation sequences presented in this work, four controlled variables were considered, i.e., the product composition A, B, C, D (X_A, $X_{B,}$ X_C, X_D). Similarly, four manipulated variables were defined, the reflux ratios (*R*) and the heat duties supplied to the reboilers (*C*). After the optimum designs were obtained, open-loop dynamic simulations were obtained in Aspen Dynamics 11.1 in order to obtain the transfer function matrix. For the case study M1F1, Tables 8–11 show some representatives transfer function matrices. It can be noted that the dynamic responses can be adjusted to first or second order models with or without dead times. Because of singular values depend on the units of the variables, it is necessary the scaling of the gains. In this work, the controlled

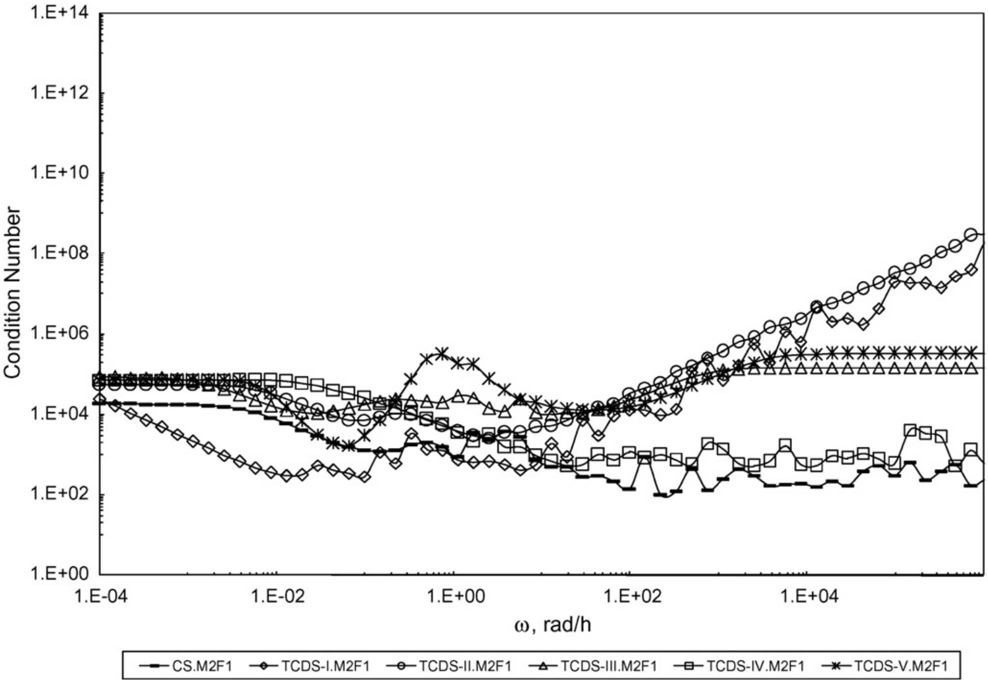


Fig. 9 – Condition numbers (M2, F1).

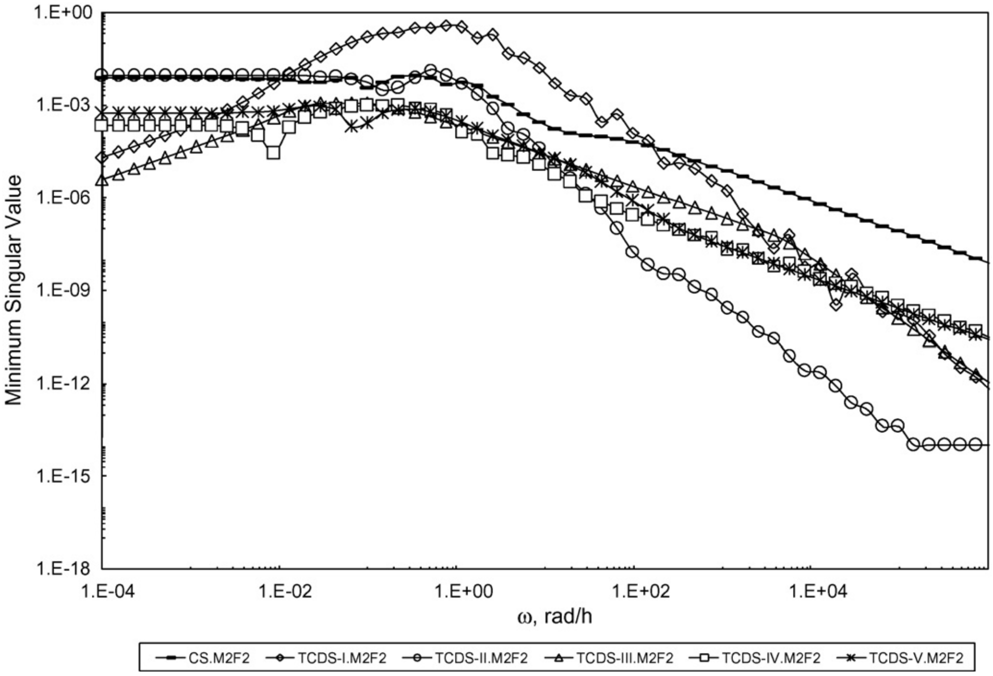


Fig. 10 – Minimum singular values (M2, F2).

variables are bounded between 0 and 1 (mole fractions) and the changes in the manipulated variables were associated to the fraction in the opening of the control valve (bounded between 0 and 1).

For the case M1F1 (Figs. 4 and 5) we got the next results at low and intermediate frequencies: the TCDS-I and the TCDS-III present, in general, lower values of the condition number and higher values of the minimum singular value; therefore, it can be expected that the TCDS-I and the TCDS-III systems exhibit better control properties than the other sequence under feedback control and they are better conditioned to the effect of disturbances than the other distillation schemes. As the frequency increases, the TCDS-II presents good dynamic behavior according to the minimum singular value (Fig. 4), but in terms of the condition number, the CDS option is better conditioned to the effect of disturbances and modeling errors. It is important to highlight that the controllability analysis conducted by singular value decomposition is a qualitative measure that can be supported with quantitative measures; for instance, the integral of the absolute error.

For the case M1F2, Figs. 6 and 7 show a clear tendency at low frequencies, i.e., the TCDS-III, TCDS-IV and TCDS-V

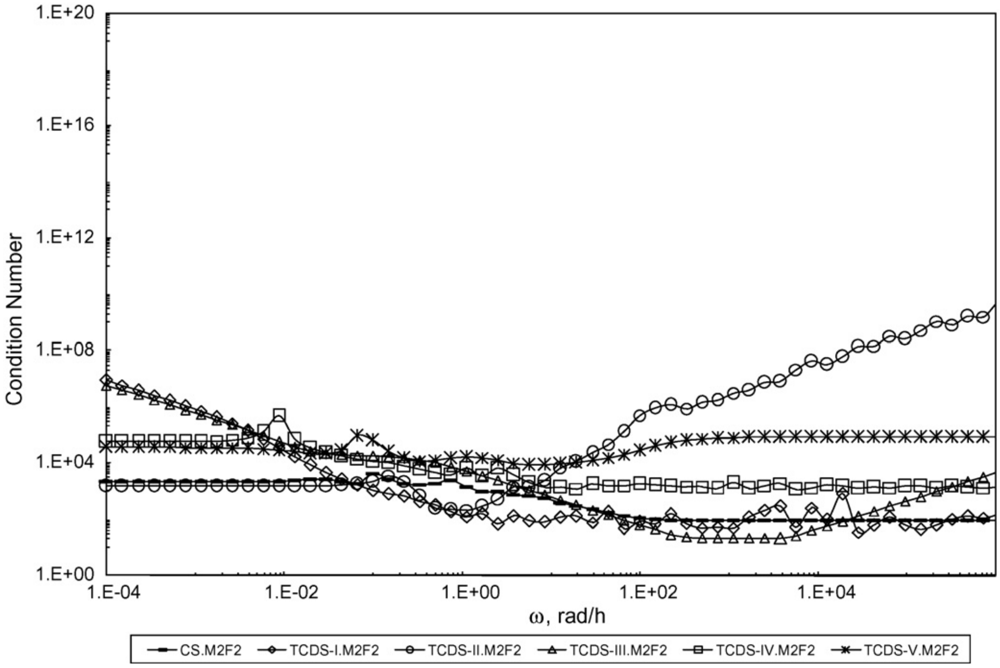
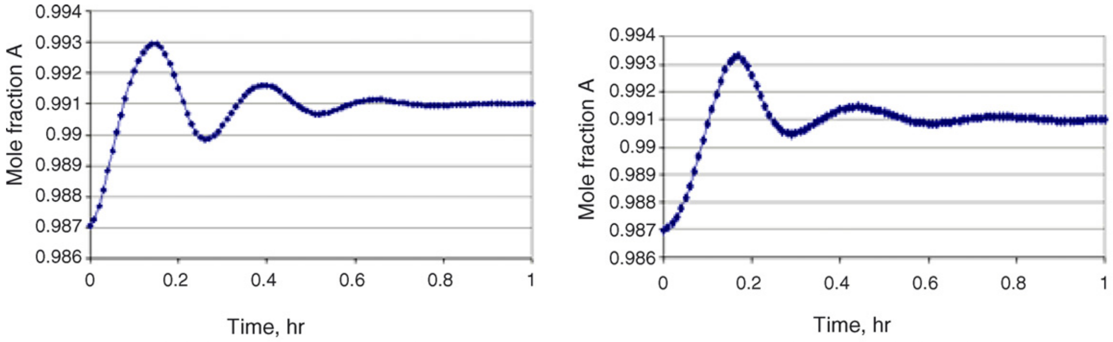
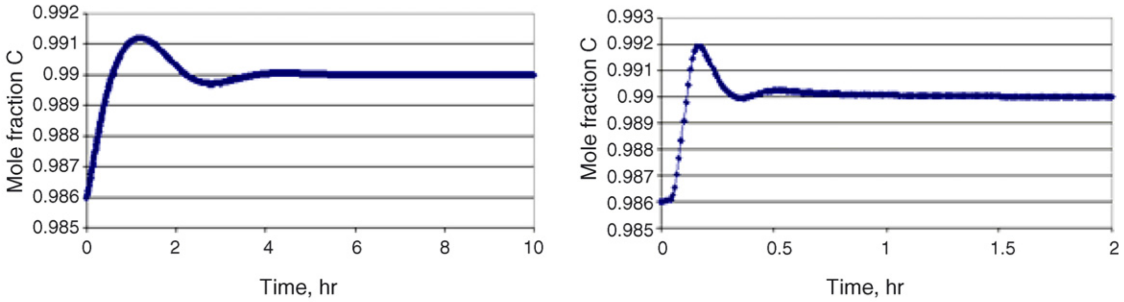


Fig. 11 – Condition numbers (M2, F2). (a) Component A (*LV* structure) (b) component A (RGA structure) (c) component C (*LV* structure) (d) component C (RGA structure).



(a) Component A (LV structure) (b) Component A (RGA structure)



(c) Component C (LV structure) (d) Component C (RGA structure)

Fig. 12 – Dynamic responses in TCDS-V for *LV* and RGA structures.

systems exhibit higher values of the minimum singular value and lower values of condition number than the other distillation sequences, but as the frequency increases, the condition number increase drastically, and the TCDS-III shows the worst values. In general, we can say that the TCDS-III, TCDS-IV and TCDS-V schemes offer better conditioning properties regarding model uncertainties and disturbances than the other arrangement at low frequencies.

Figs. 8 and 9 display the minimum singular values and the condition numbers respectively, for the case M2F1. The TCDS-II and TCDS-IV schemes have the highest values of condition number and the lowest values of the σ_* for the

Table 12 – IAE results for mixture M1F1 (*LV* control structure)

Sequence	Component	K_C	τ_I	IAE
CS	A	254	64	1.9899E-04
	B	15	33	1.7569E-04
	C	256	48	5.5710E-05
	D	500	1	1.4323E-05
TCDS-I	A	200	48	1.49655E-05
	B	480	37	6.9105E-01
	C	200	92	1.60134E-05
	D	200	2	2.92458E-05
TCDS-II	A	200	54	2.0030E-04
	B	200	4	1.8508E-04
	C	19	67	5.2000E-04
	D	200	51	6.7799E-05
TCDS-III	A	200	67	2.11189E-04
	B	200	39	6.59889E-05
	C	350	55	9.00541E-01
	D	200	2	3.23525E-05
TCDS-IV	A	200	67	2.1734E-04
	B	200	41	7.0714E-05
	C	99	130	1.2783E-04
	D	200	62	4.0691E-05
TCDS-V	A	200	11	1.7958E-04
	B	450	14	3.8874E-01
	C	490	8	8.9567E-01
	D	100	3	5.1897E-05

Table 13 – IAE results for mixture M2F1 (*LV* control structure)

Sequence	Component	K_C	τ_I	IAE
CS	A	200	87	3.02098E-04
	B	200	53	1.05494E-04
	C	3	958	4.83194E-03
	D	200	9	1.50155E-05
TCDS-I	A	200	104	1.91234E-04
	B	410	100	9.87510E-01
	C	24	200	5.21765E-05
	D	200	9	1.50428E-05
TCDS-II	A	200	110	2.1694E-04
	B	200	5	2.8507E-04
	C	2	1019	7.8109E-03
	D	200	9	1.4935E-04
TCDS-III	A	200	90	3.01807E-04
	B	200	58	9.93708E-05
	C	64	200	2.99117E-04
	D	67	9	3.16064E-04
TCDS-IV	A	200	80	3.0412E-04
	B	200	56	1.9810E-04
	C	4	1623	5.9355E-03
	D	200	7	1.3606E-04
TCDS-V	A	200	180	2.89582E-04
	B	500	89	4.0014E-01
	C	390	140	8.87622E-01
	D	13	22	1.21517E-03

Table 14 – IAE results for mixture M2F1 (*LV* control structure and structure proposed by RGA)

Sequence	Component	IAE (*LV* structure)	IAE (structure proposed by RGA)
CS	A	1.9899E-04	5.6832E-04
	B	1.7569E-04	1.7059E-04
	C	5.5710E-05	4.9982E-03
	D	1.4323E-05	6.4705E-04
TCDS-V	A	1.7958E-04	1.7958E-05
	B	3.8874E-01	2.50004E-01
	C	8.9567E-01	6.60134E-04
	D	5.1897E-05	6.8353E-04

whole frequency range when it is compared with all sequences. At low and moderate frequencies, Fig. 8 shows higher values of the minimum singular value and Fig. 9 presents lower values of the condition number for the TCDS-I option. As a result, it is expected that this complex distillation scheme will exhibit good dynamic properties in comparison to other schemes.

The results for M2F2 are displayed in Figs. 10 and 11. At intermediate frequencies the TCDS-I shows the best results (lower condition number and higher minimum singular value). The TCDS-II and the CS systems exhibit good results at low frequencies. As a result, it can be expected that thermally coupled distillation systems are better conditioned to the effect of disturbances than the other configurations.

Similar results were obtained in the case of mixture M3. We can see, in general, that CS does not show the worst results in comparison with other schemes. As a result, it can be expected that thermally coupled distillation systems without change of structural topology are better conditioned to the effect of disturbances than the conventional arrangements. The change of the topology affects the control properties (TCDS-II and TCDS-IV) of the complex arrangements.

4.3. *Dynamic results*

To supplement the SVD analysis, rigorous dynamic simulations under closed loop operation were carried out. 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 (see for instance Häggblom and Waller, 1992). 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. Two control loops arise naturally from the experience on the operation of conventional columns. For the control of product, when it is obtained as an overhead product, the reflux flowrate was used; for the control of product, when it is obtained as a bottom product, the reboiler heat duty was chosen (see Fig. 3). 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 IAE criterion. For the dynamic analysis, individual set point changes for product composition were implemented for each of the four product streams (as showed in Fig. 12). For all cases (conventional and integrated sequences), the four 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.

Table 12 shows the IAE values obtained for each composition control loop of the six cases for mixture M1, when feed F1 was considered. The TCDS-I and the TCDS-III offered the best dynamic behavior, based on the lowest values of IAE, for the control of the four product streams. This result is in agreement with that obtained in the SVD analysis. In Table 13 are displayed the IAE results for the case M2F1. TCDS-I exhibits, in general, the lower IAE values in comparison with other sequences. Again, this result supports that obtained in the SVD analysis.

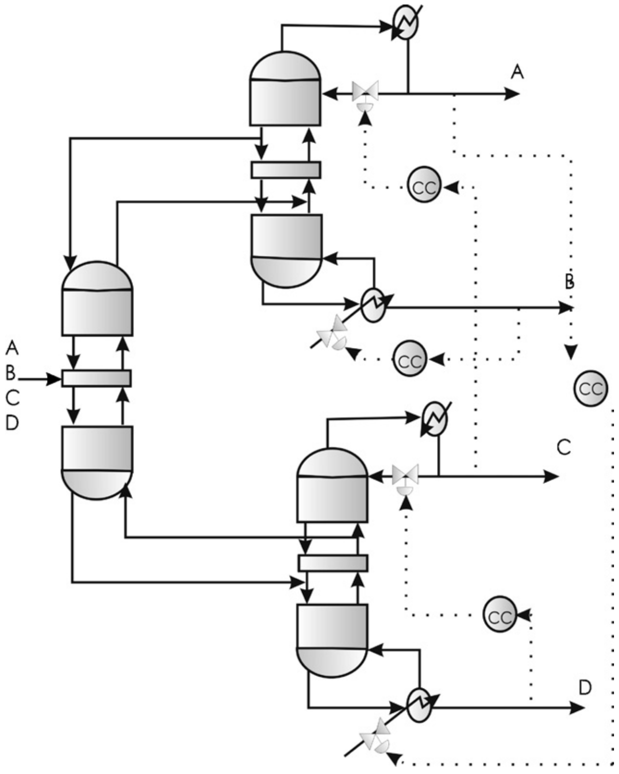


Fig. 13 – Control loops proposed by RGA method for the TCDS-V scheme.

To complete the study of the control properties, relative gain arrays (RGA) were obtained for the six distillation sequences. Matrix 1 shows the RGA for the TCDS-V. As we can see, the complex sequence presents interactions because positive values of the relative gains are greater than 1. Also, the negative values cause negative interactions.

$$\Lambda = \begin{pmatrix} 6.787137443 & -5.973190315 & -1.292531039 & 1.478583911 \\ -10.87000059 & 10.00274336 & -3.326438693 & 5.193695919 \\ 0.891530136 & 1.928236382 & -0.671878701 & -1.147887817 \\ 4.191333011 & -4.95778943 & 6.290848433 & -4.524392013 \end{pmatrix} \quad (1)$$

In general, from relative gains arrays for all cases of study, we can conclude that significant interactions are present in both conventional and thermally coupled distillation sequences. We use the relative gain array method to fix other loops for the control system. It is important to note that we have conducted the RGA analysis in the low frequency region. Moreover, this consideration will most likely give meaningless results if there are large relative gains within the bandwidth where the model is going to be used (Skogestad and Postlethwaite, 2005).

One representative case is displayed in Table 14. We show the IAE values for both cases (*LV* and RGA control structure). The structure proposed by RGA is showed in Fig. 13 for the TCDS-V structure. The IAE values for RGA structure, in the case of complex sequence, are better than *LV* control loops (see Fig. 12). The pairing proposed by RGA is better in the case of TCDS-V.

5. Conclusions

We have conducted a comparison on the energy consumption, η, TAC and dynamic behavior of five complex distillation sequences for the separation of quaternary mixtures. One factor that seems to affect the optimal choice is the structure of the complex sequence. Schemes with thermally couplings present the best values of energy consumption, η and TAC in contrast to the conventional distillation sequences. However, if the schemes do not have change in their topology, they can show better dynamic behavior (TCDS-I and TCDS-III show the best results in both the SVD and closed loop dynamic analysis). In summary, although the best operational option is not unique, the results show that there are cases in which integrated sequences do not only provide significant energy savings with respect to the conventional sequences, but also may offer some dynamic advantages.

Acknowledgements

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Appendix A

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

First Law of the thermodynamic:

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

Second Law of the thermodynamic:

$$\sum_{\text{out of system}} \left(ns + \frac{Q}{T_s}\right) - \sum_{\text{in to system}} \left(ns + \frac{Q}{T_s}\right) = \Delta S_{irr} \quad \text{(A.2)}$$

Exergy balance:

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

Minimum work of separation:

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

Second law efficiency:

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

where $b = h - T_0 s$ is the exergy function, $LW = T_0 \Delta S_{irr}$ is the lost work in the system and $\boldsymbol{\eta}$ is the thermodynamic efficiency. The thermodynamic properties like enthalpies and entropies of the streams of the distillation sequences were evaluated through the use of the simulator of processes Aspen Plus 11.1™.

Appendix B

For a given number of theoretical trays, Aspen Plus simulator calculates column diameter and height (for 24 in tray spacing) after converging for selected valve tray column with 2 in weir height. Valve trays of Glitsch type are considered. The costing of distillation column (carbon steel construction) was estimated by the cost equations showed in Turton et al. (2004) that are updated with the CEPCI (Chemical Engineering Process Cost Index). For comparison a single value of CEPCI = 473.10 is selected (October, 2005), value of the starting the year of this research. The total column cost is the sum of the installed cost of column shell and the installed cost of column trays. On the other hand, the sizing and costing of heat exchangers were calculated. The cost of heat exchangers can be correlated as a function of the surface area assuming shell and tube, floating head, and carbon steel construction. The installation prices are updated by the CEPCI index. The capital cost (purchase plus installation cost) is annualized over a period, which is often referred to as plant life time:

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

$$\begin{aligned} \text{Total annual cost(TAC)} &= \text{Annual operating cost} \\ &\quad + \text{Annual capital cost} \end{aligned} \quad \text{(B.2)}$$

Operating costs were assumed just utility cost (steam and cooling water).

Plant life = 5 years

Operating hours = 8400 h/year.

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