# Dynamic Behavior of Thermally Coupled Distillation Configurations for the Separation of Multicomponent Mixtures

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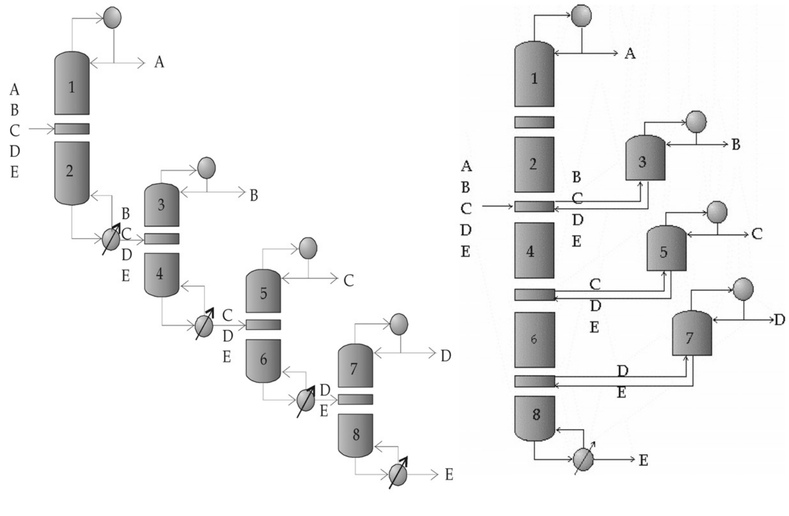
Original scientific paper
Received: October 4, 2005
Accepted: March 15, 2006

Control properties of thermally coupled distillation arrangements for the separation of multicomponent mixtures were compared to those of conventional distillation sequences. Seven thermally coupled schemes were investigated. The preliminary steady – state design of complex schemes was obtained by starting from a conventional distillation sequences and then optimizing for minimum energy consumption (energy-efficient designs). The dynamic behavior of the sequences considered were obtained by using the singular value decomposition technique at zero frequency. It was found that, in general, the complex schemes present theoretical control properties similar or better to those of conventional distillation sequences. This result is significant because it lets one establish that the energy savings predicted for thermally coupled distillation sequences are achieved without introducing additional control problems.

*Key words:*
Thermally coupled distillation sequences, energy requirements, dynamic properties, multicomponent mixtures

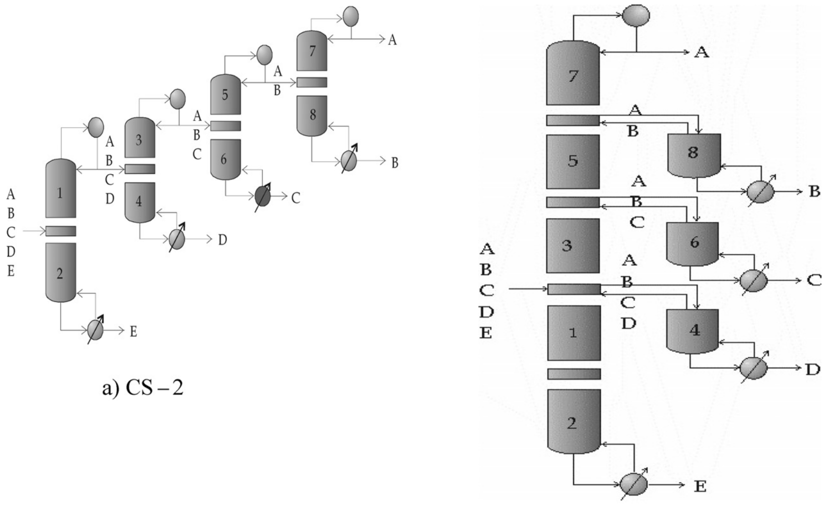
## Introduction

Distillation processes in petroleum refineries consume a significant portion of the energy demand of the site, in spite of extensive heat recovery. Heat integration has proven to be successful in reducing the energy costs for conventional distillation arrangements. However, the scope for integrating conventional distillation columns is often limited. Such limitations call for non-conventional column designs to be considered. One of the most important non-conventional distillation arrangements involves thermal coupling. The use of thermal coupling has until recently been almost exclusively restricted to side-strippers in petroleum industry. It has been established that energy savings of 30 % are typical when thermally coupled schemes are compared with a conventional arrangement. However, due to the complexity, research on these complex distillation configurations is only restricted to three – component mixtures, for only a few promising flowsheets can be constructed for ternary mixtures (*Tedder* and *Rudd*[1]; *Glinos* and *Malone*[2]). Much detail work has been contributed on some specific ternary configurations aiming at the performance analysis (*Carlberg* and *Westerberg*;[3] *Hernández* and *Jiménez*;[4] *Hernández* and *Jiménez*;[5] *Yeomans* and *Grossman*;[6] *Rev* et al.[7]). Also, the thermally coupled distillation sequences for the separation of ternary mixtures, over a wide range of relative volatilities and feed compositions, have been reported to provide a better thermodynamic efficiency than the conventional distillation configurations (*Flores* et al.[8]). For mixtures with four or more components, the combinatorial problem makes it even more difficult to construct all the possible schemes. There are few works on extensions toward the design of integrated systems for mixtures of more than three components (*Agrawal*;[9] *Christiansen* et al.;[10] *Blancarte-Palacios* et al.;[11] *Rong* et al[12,13,14]). Recently, *Calzon – McConville* et al.[15] have parametrically studied some thermally coupled distillation sequences for five – component mixtures from viewpoint of energy consumption. In general, the economical potential of thermally coupled sequences has already been recognized, but their control properties have not been studied to the same degree. Recent efforts have contributed to the understanding of the dynamic properties of integrated schemes for the separation of ternary and quaternary mixtures (*Abdul – Mutalib* and *Smith*;[16] *Hernández* and *Jiménez*;[17] *Jiménez* et al.;[18] *Segovia – Hernández* et al.;[19] *Segovia – Hernández* et al.;[20] *Segovia – Hernández* et al.;[21] *Cardenas* et al.[22]). The expectance that the dynamic properties of those coupled systems may cause more operational problems than the conventional sequences is one of the factors that has contributed to their lack of industrial implementation. This conflict is commonly observed in cases where the optimization of an energy – efficient systems leads to tight designs, which in turn are more difficult to control. In this work we developed a comparative study of the control properties of the seven thermally coupled distillation sequences (previously studied by *Calzon – McConville* et al.[15]) for the separations of five – component mixtures to those of conventional sequences (Fig 1 – 7).



a) CS − 1 b) TCDS − 1

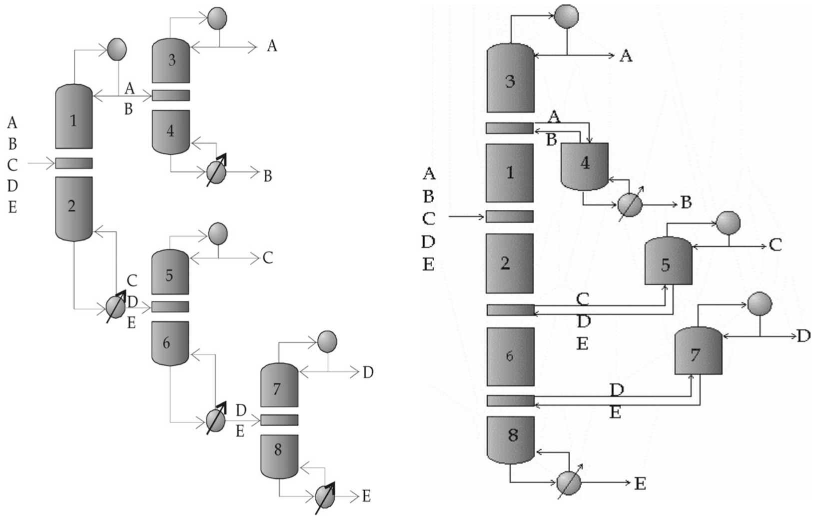
Fig. 1 – *Sequence fully thermally coupled (TCDS – 1) and the conventional sequence where it is obtained*



a) CS – 2

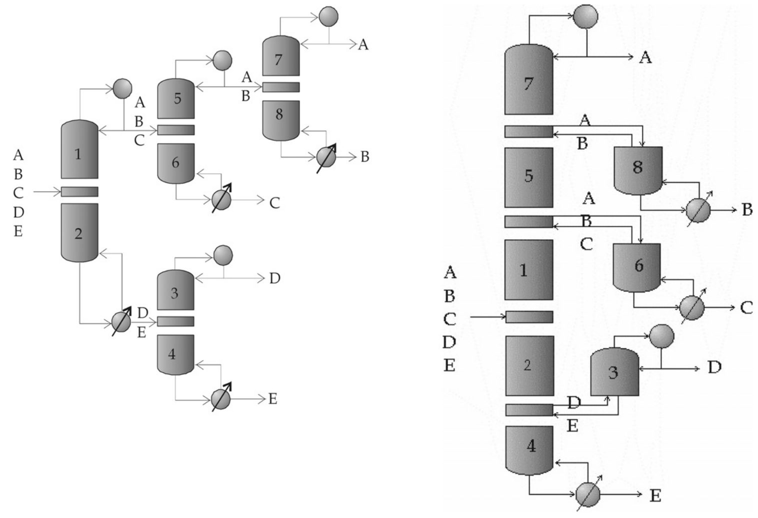
b) TCDS – 2

Fig. 2 – *Sequence fully thermally coupled (TCDS – 2) and the conventional sequence where it is obtained*



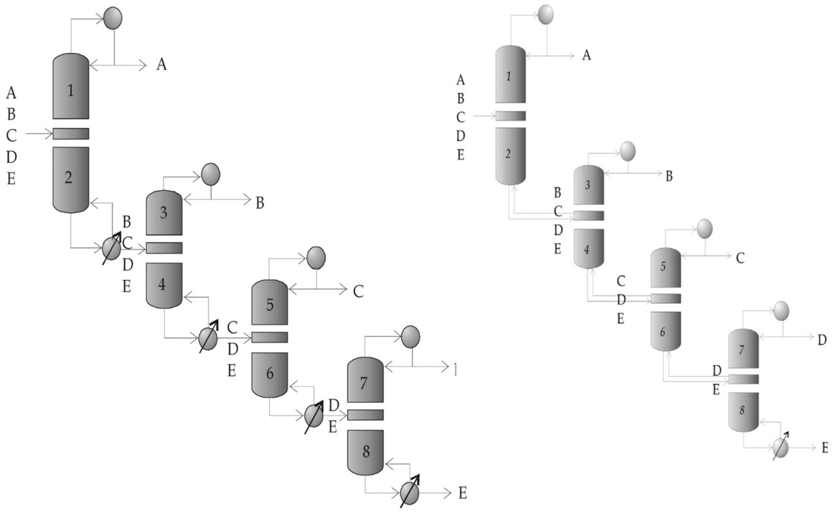
a) CS – 3 b) TCDS – 3

Fig. 3 – *Sequence fully thermally coupled (TCDS – 3) and the conventional sequence where it is obtained*



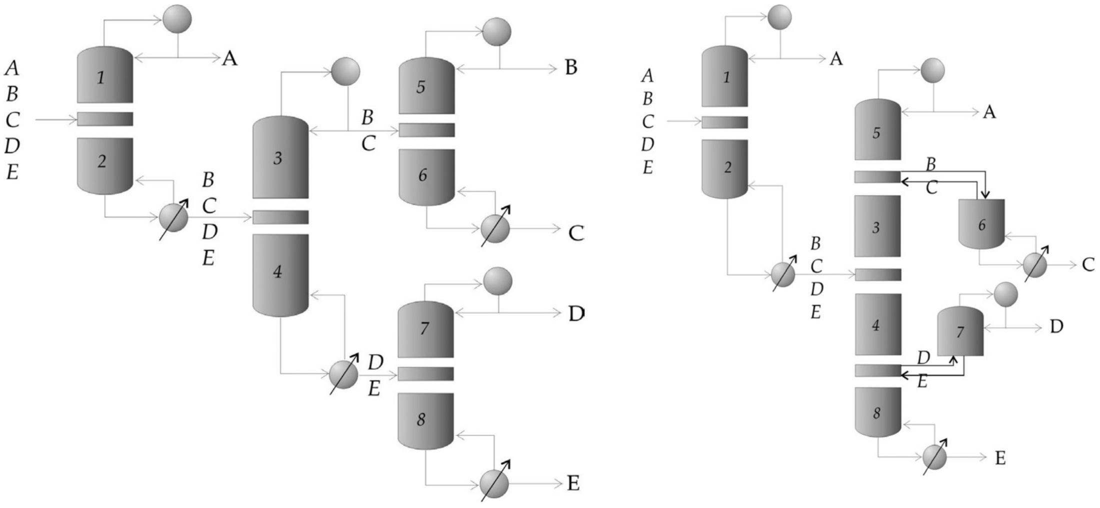
a) CS – 4 b) TCDS – 4

F i g . 4 – *Sequence fully thermally coupled (TCDS – 4) and the conventional sequence where it is obtained*



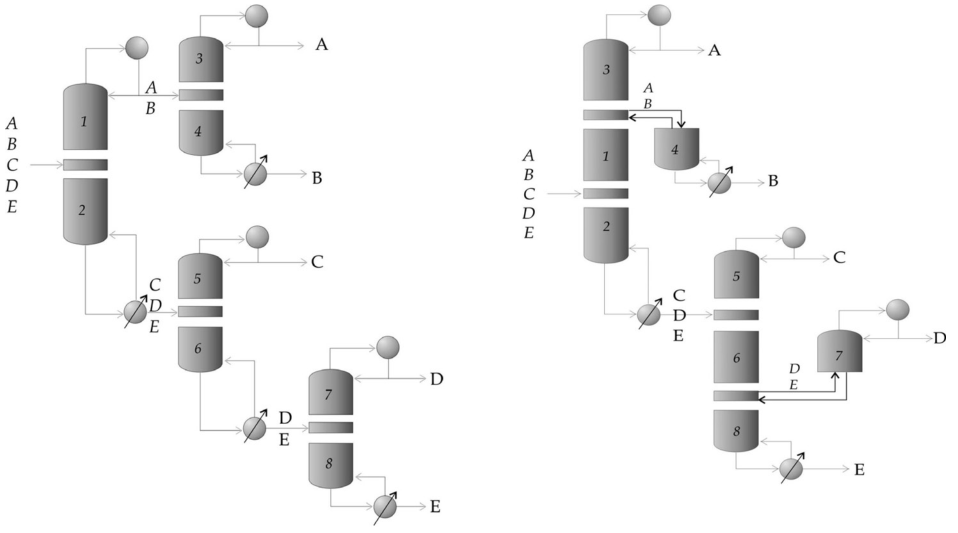
a) CS – 5 b) TCDS – 5

F i g . 5 – *Sequence fully thermally coupled (TCDS – 5) and the conventional sequence where it is obtained*



a) CS – 6 b) TCDS – 6

F i g . 6 – *Sequence partially thermally coupled (TCDS – 6) and the conventional sequence where it is obtained*



a) CS – 7 b) TCDS – 7

Fig. 7 – *Sequence partially thermally coupled (arrangement TCDS – 7) and the conventional sequence where it is obtained*

## Design of complex schemes

Strictly, the design of the thermally coupled distillation sequences could be modeled through superstructures suitable for optimization procedures with mathematical programming techniques. However, the task is complicated and is likely to fail to achieve convergence. 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 stage of our approach begins with the development of preliminary designs for the complex systems from the design aspects on conventional distillation columns (See Fig. 1 – 7). In the conventional sequences, each column performs its respective split (i.e. the separation of the light and heavy key components) with a recovery of 98 %. Using the well – know short cut methods of Fenske – Underwood and Gilliland, 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.

The conventional sequences (CS; Fig. 1 – 7) show eight different tray sections. These sections are used as a basis for the arrangement of the tray structure of the coupled schemes through a section analogy procedure. For instance, in the main column of the integrated sequence of Fig. 1, the total number of trays is obtained by conceptually moving stripper sections from the second to four column of the conventional sequence to the bottom of the first column. The reboiler of the first column is replaced by a vapor –liquid interconnection with a side rectifier. The number of trays in the side rectifiers of the complex scheme is equal to the number of trays in the rectifier zone in the second, third and fourth column (in the conventional arrangement), respectively. Two more vapor –liquid interconnections are used to get the coupling scheme. A similar procedure is applied to obtain the other thermally coupled schemes.

After the tray arrangement for the integrated designs has been obtained, an optimization procedure is used to minimize the heat duty supplied to the reboilers of each coupled scheme, taking into account the constrains imposed by the required purity of the five products streams. Although, the number of trays is not formally optimized a parametric analysis can be carried out to test different tray arrangements by changing the recoveries of the key components. Depending the recovery on the key components, the number of trays changes. Because the resulting sections serve as a basis for the tray arrangements of the complex scheme, such a procedure allows for the comparison of different designs to detect the design with superior performance in terms of energy consumption. In practice, the procedure is limited by the number of tray arrangements that the designer decides to consider.

Then, the degrees of freedom that remain, after design specifications and tray arrangement are used to obtain the operating conditions, are used to get the integrated designs which provide minimum energy consumption. Three or two degrees of freedom (depending if the scheme is fully thermally coupled or partially thermally coupled. See Fig. 1 – 7) remain for each integrated sequence. They are the interconnecting flows (vapor or liquid, depending on the scheme). The search procedure provides the optimal values of the interconnecting flow to minimize the energy consumption for the separation. The design is successful if it meets the product specifications. The optimization strategy is described in Fig. 8 (more details about the procedure are in *Calzon – McConville* et al.[15]).

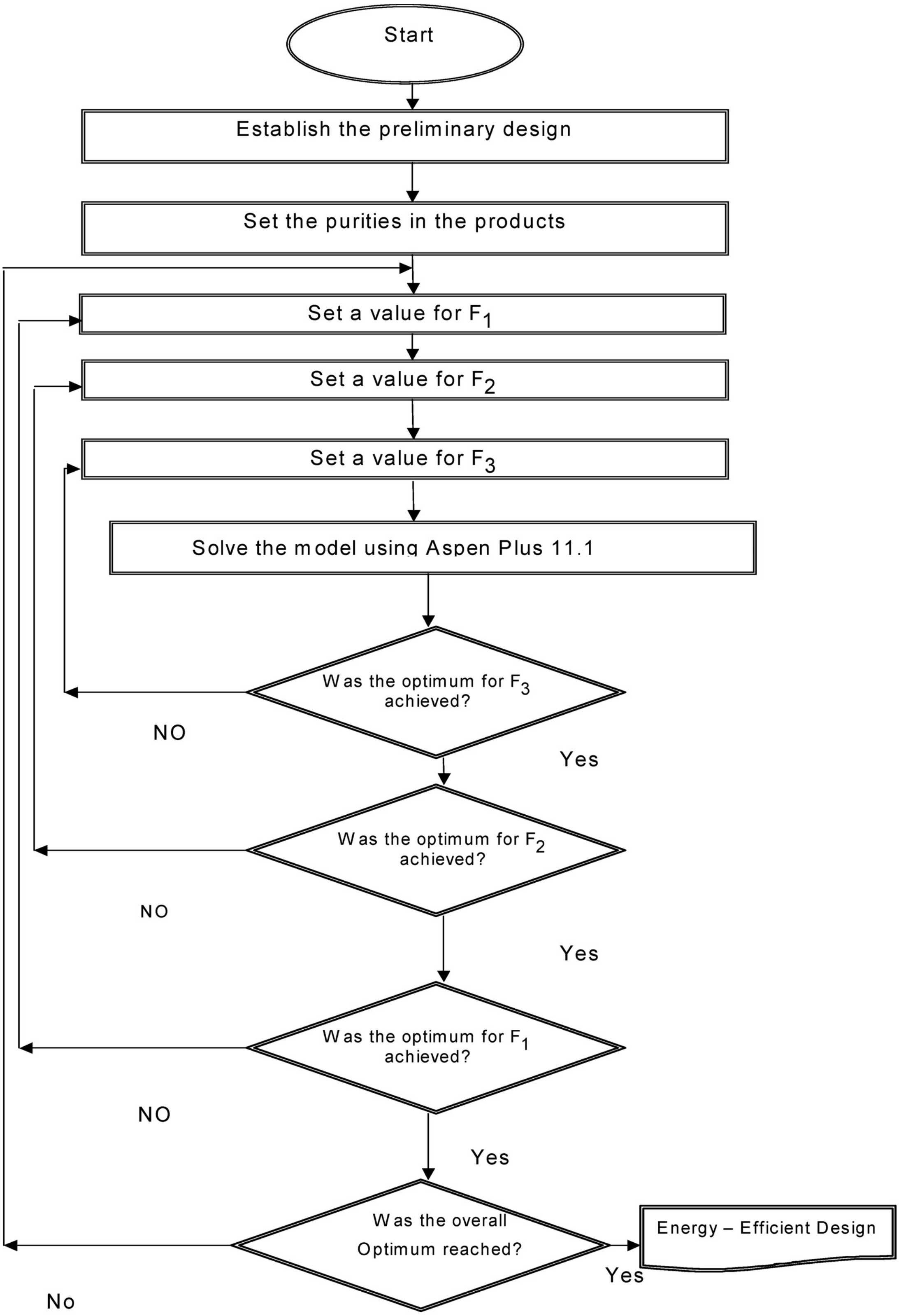


Fig. 8 – *Strategy to get the energy efficient design*

## Control properties

Open loop dynamic responses to set point changes 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. Transfer function matrices ($\boldsymbol{G}$) were then collected for each case, and they were subjected to singular value decomposition (SVD):

$$\boldsymbol{G} = \boldsymbol{V}\,\Sigma\,\boldsymbol{W}^{\mathrm{H}} \tag{1}$$

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

$$\gamma = \sigma^* \,/\, \sigma_* \tag{2}$$

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 (*Papastathopoulou* and *Luyben*[23]). A full SVD analysis should cover a sufficiently complete range of frequencies. For this initial analysis of the coupled schemes to the conventional configurations, we simply estimated the SVD properties for each separation system at zero frequency. Such analysis should give some preliminary indication on the control properties of each system around the nominal operating point.

## Case of study

The analysis presented in this work is based on the separation problem of two different five – component mixtures with molar compositions (A, B, C, D, E) equal to (0.35, 0.10, 0.10, 0.10, 0.35; $F_1$) and (0.125, 0.25, 0.25, 0.25, 0.125; $F_2$), to examine the effect of the content of the intermediate components, and product purities of 98, 94, 94, 94 and 97 %, respectively. The two mixtures considered were n-butane, n-pentane, n-hexane, n-heptane and n-hexane (mixture 1), n-butane, isopentane, n-pentane, n-hexane and n-heptane (mixture 2). A feed flowrate of 45.5 kmol h$^{-1}$ as saturated liquid was taken. The design pressure for each sequence was chosen such that all condensers could be operated with cooling water. The pressure drop for a single tray is given based on the heuristics of *Kister.*[24] Since the feed involves a hydrocarbon mixture, the Chao – Seader correlation was used for the prediction of thermodynamic properties (*Seader* and *Henley*[25]). The tray arrangements and some parameters for the TCDS – 1, after optimization task for mixture 1 ($F_1$), is given in Tab. 1. It is important to establish that studying a five – component mixture of hydrocarbons is a suitable example, given the applications of the hydrocarbon mixtures in the petrochemical industry (*Harmsen*[26]). As far as energy consumption is concerned, the optimized steady – state design provides energy savings of ~35 % with respect to the best energy – efficient sequence based on conventional distillation columns (Tab. 2 and 3; more details about energy consumption in *Calzon – McConville* et al.[15]). During the search for the optimum energy consumption, five design specifications for products purities (A, B, C, D, E) were set in order to avoid deviations in the required purities.

Table 1 – *Sequence design for the TCDS–1, mixture 1 (F1)*

| Column | Quantities |
|---|---|
| Main column | stages = 41<br>feed stage = 9<br>reflux ratio = 1.72<br>$F_{\mathrm{V1}}$ = 16.83 kmol h$^{-1}$<br>$F_{\mathrm{V2}}$ = 20 kmol h$^{-1}$<br>$F_{\mathrm{V3}}$ = 22.75 kmol h$^{-1}$<br>pressure = 4.5 bar |
| Side rectifier 1 (where component B is purified) | stages = 10<br>distillate flowrate = 4.6 kmol h$^{-1}$ |
| Side rectifier 2 (where component C is purified) | stages = 11<br>distillate flowrate = 4.45 kmol h$^{-1}$ |
| Side rectifier 3 (where component D is purified) | stages = 11<br>distillate flowrate = 4.54 kmol h$^{-1}$ |

## Results

The theoretical control properties of conventional and thermally coupled distillation sequences were obtained. 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 (base designs) and registering the dynamic responses of the five products. Open – loop dynamic simulations were carried out in Aspen Dynamic 11. 1 in order to obtain the transfer function matrix.

Table 2 – *Optimum energy requirements (kW) for fully thermally coupled sequences, mixture 1*

| Feed | Sequence | Total reboiler duty (kW) |
|---|---|---|
| $F_1$ | CS – 1 | 1090 |
| | TCDS – 1 | 841 |
| | CS – 2 | 1181 |
| | TCDS – 2 | 793 |
| | CS – 3 | 1062 |
| | TCDS – 3 | 931 |
| | CS – 4 | 1168 |
| | TCDS – 4 | 708 |
| | CS – 5 | 1092 |
| | TCDS – 5 | 841 |
| $F_2$ | CS – 1 | 2032 |
| | TCDS – 1 | 1246 |
| | CS – 2 | 2039 |
| | TCDS – 2 | 1471 |
| | CS – 3 | 1213 |
| | TCDS – 3 | 1123 |
| | CS – 4 | 1332 |
| | TCDS – 4 | 960 |
| | CS – 5 | 2032 |
| | TCDS – 5 | 1246 |

Table 3 – *Optimum energy requirements (kW) for partially thermally coupled sequences, mixture 1*

| Feed | Sequence | Total reboiler duty (kW) |
|---|---|---|
| $F_1$ | CS – 6 | 1446 |
| | TCDS – 6 | 1157 |
| | CS – 7 | 1484 |
| | TCDS – 7 | 1096 |
| $F_2$ | CS – 6 | 1222 |
| | TCDS – 6 | 960 |
| | CS – 7 | 1482 |
| | TCDS – 7 | 1241 |

Table 4 – *Minimum singular value and condition number for sequences fully thermally coupled in the mixture 1*

| Feed | Sequence | $\sigma_*$ | $\gamma$ |
|---|---|---|---|
| $F_1$ | CS – 1 | 0.0219 | 562 |
| | TCDS – 1 | 0.0218 | 564 |
| | CS – 2 | 0.0012 | 14106 |
| | TCDS – 2 | 0.2026 | 33 |
| | CS – 3 | 0.0005 | 33318 |
| | TCDS – 3 | 0.0418 | 576 |
| | CS – 4 | 0.0064 | 59934 |
| | TCDS – 4 | 0.0071 | 56650 |
| | CS – 5 | 0.0350 | 562 |
| | TCDS – 5 | 0.0001 | 14000 |
| $F_2$ | CS – 1 | 0.0401 | 490 |
| | TCDS – 1 | 0.0405 | 488 |
| | CS – 2 | 0.0176 | 109 |
| | TCDS – 2 | 0.0560 | 14 |
| | CS – 3 | 0.0008 | 10007 |
| | TCDS – 3 | 0.0988 | 689 |
| | CS – 4 | 0.0111 | 40098 |
| | TCDS – 4 | 0.0998 | 4765 |
| | CS – 5 | 0.0500 | 129 |
| | TCDS – 5 | 0.0009 | 9000 |

Table 5 – *Minimum singular value and condition number for sequences fully thermally coupled in the mixture 2*

| Feed | Sequence | $\sigma_*$ | $\gamma$ |
|---|---|---|---|
| $F_1$ | CS – 1 | 0.0906 | 601 |
| | TCDS – 1 | 0.0900 | 606 |
| | CS – 2 | 0.0456 | 309 |
| | TCDS – 2 | 0.0908 | 35 |
| | CS – 3 | 0.0007 | 9005 |
| | TCDS – 3 | 0.0605 | 977 |
| | CS – 4 | 0.0028 | 43982 |
| | TCDS – 4 | 0.0109 | 1008 |
| | CS – 5 | 0.0621 | 908 |
| | TCDS – 5 | 0.0003 | 7406 |
| $F_2$ | CS – 1 | 0.0821 | 376 |
| | TCDS – 1 | 0.0829 | 369 |
| | CS – 2 | 0.0198 | 469 |
| | TCDS – 2 | 0.0576 | 90 |
| | CS – 3 | 0.0004 | 7005 |
| | TCDS – 3 | 0.0129 | 884 |
| | CS – 4 | 0.0060 | 28065 |
| | TCDS – 4 | 0.0281 | 2974 |
| | CS – 5 | 0.0807 | 1004 |
| | TCDS – 5 | 0.0004 | 12754 |

### Sequences fully thermally coupled (three thermally couplings)

Table 4 and 5 give the results for the SVD test for each sequence. In the case of TCDS – 1 has similar values for minimum singular value and condition number than conventional sequence (CS – 1), which implies that the coupled arrangement has similar control properties (lower control efforts under feedback operation and better conditioned to the effect of disturbances) than the conventional design. In the case of TCDS – 2, TCDS – 3, TCDS – 4 present higher values of the minimum singular value (Tab. 4 and 5); therefore, it can be expected that that these coupled systems exhibit better control properties than the conventional sequences under feedback control. The results for the condition number show that the complex sequences offer the best values (Tab. 4 and 5). As a result, it can be expected that thermally coupled distillation systems are better conditioned to the effect of disturbances than the conventional arrangements.

One detail is worth highlighting, the TCDS – 1 and TCDS – 5 are thermodynamic equivalents schemes. A simple column has two sections, the rectifying and stripping sections. Based on the function of each of the column sections in a complex distillation schemes (i.e., either rectifying sections or stripping sections), a complex scheme can be converted into a sequence in which each unit has only one rectifying column section and one stripping column section. Then the connections of the units are determined according to the interconnections of their streams. Thus, for the TCDS – 1 scheme, TCDS – 5 could be obtained. That converted sequence is a thermodynamic equivalent scheme of the corresponding TCDS – 1 (*Carlberg* and *Westerberg*[3]). The results (Tab 4 and 5) show that TCDS – 5 have the worst control properties in comparison with the TCDS 1 – 4. In this case the conventional scheme (CS – 5) have better values in the minimum singular value and condition number. This result is important because two thermodynamically equivalent schemes have different dynamic properties. In other words, the structure has different effect to dynamic performance for a specific separation. This situation can be analyzed in this form: the structure with side columns have better control properties than a thermodynamic scheme with retrofit. Other results about this topic can be observed in *Santos – Méndez* and *Hernández*[27] in the case of quaternary mixtures.

### Sequences partially thermally coupled (two thermal couplings)

For the case of sequences partially thermally coupled (structures with reduction in thermal couplings) the results are presented in Tab. 6. In both cases analyzed the complex schemes show higher values of the minimum singular value and offer the best values in the condition number. Therefore, it can be expected that these coupled systems exhibit better control properties than the conventional sequences under feedback control and, it can be expected that systems are better conditioned to the effect of disturbances than the conventional arrangements.

Table 6 – *Minimum singular value and condition number for sequences partially thermally coupled in the mixture 1*

| Feed | Sequence | $\sigma_*$ | $\gamma$ |
|---|---|---|---|
| $F_1$ | CS – 6 | 0.00220 | 13588 |
| | TCDS – 6 | 0.00500 | 2630 |
| | CS – 7 | 0.00003 | 19004 |
| | TCDS – 7 | 0.00009 | 1974 |
| $F_2$ | CS – 6 | 0.00010 | 21349 |
| | TCDS – 6 | 0.00024 | 9264 |
| | CS – 7 | 0.00006 | 34450 |
| | TCDS – 7 | 0.00079 | 3884 |

A remark on partially thermally coupled structures can be established. Some authors (for instance, *Agrawal* and *Fidkowski*[28,29]) have claimed that the reduction of interconnection flows in complex arrangements might provide better operating properties. When we do a general comparison of control properties of the seven coupled schemes (Tab. 4 – 6), the partially thermally coupled (TCDS 6 – 7) arrangements show worst values (minimum singular value and condition number) than the fully thermally coupled schemes (TCDS 1 – 4). This observation is interesting because the number of the thermal links affect the control properties of coupled systems. In this case, the control properties have been improved with the presence of three recycles.

## Conclusions

Controllability properties of seven thermally coupled distillation sequences for the separation of five component mixtures have been shown to be better than those of the conventional sequences (the integrated sequence present similar or better minimum singular values and condition number in comparison to those obtained in the conventional distillation sequences for both mixtures analyzed). The results show that the structure has different effect to dynamic performance for a specific separation in the case of thermodynamically equivalent schemes. These results are significant because they let us establish that coupled schemes not only require energy demands lower than the conventional distillation sequences but also present theoretical control properties similar or better to those of the conventional distillation sequences used in the preliminary design of the thermally coupled distillation sequences. Similar results have been obtained by *Hernández* and *Jiménez*[17] and *Segovia – Hernández* et al.[30] for the separation of ternary mixtures and *Cárdenas* et al.[22] for the separation of quaternary mixtures. In general, it is apparent that the presence of recycle streams instead of deteriorating the dynamic behavior of thermally coupled distillation sequences (for the separation of "*n*" components), may contribute positively to their dynamic properties.

*ACKNOWLEDGMENT*

*The authors acknowledge financial support received from PROMEP and Universidad de Guanajuato, México.*

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