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Effects of intensification on process features and control properties of lignocellulosic bioethanol separation and dehydration systems

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ABSTRACT

Separation and dehydration process is a key step to reduce the total production cost of lignocellulosic bioethanol. In the earlier work (Torres-Ortega and Rong, 2016), we have obtained new intensified systems for lignocellulosic bioethanol separation and dehydration through dividing wall columns, which have considerable reduction to both capital and energy costs. This work presents the analysis of process features and control properties of the intensified systems with similar capital reduction and energy savings. The control properties were based on singular value decomposition (SVD) and dynamic performances under mild disturbances and changes of set point in Aspen Dynamics V8.8. The control properties and dynamic responses of the intensified separation systems were examined against the reference system for their structural changes during intensification by thermal couplings and column section recombination. The simultaneous analysis of process feature changes by intensification and their control properties achieved the intensified systems with both cost savings and competitive control properties.

1. Introduction

Separation and dehydration of lignocellulosic bioethanol typically starts from a fermentation broth with 5 wt. % of bioethanol, and a mixture of water, soluble organic matter, gases and insoluble solids. Once bioethanol is concentrated, it needs to be dehydrated to a purity of 99.5 wt %. However, a bioethanol-water azeotrope (95.63 wt. % bioethanol) hinders the use of conventional distillation.

Regarding separation and dehydration of lignocellulosic bioethanol, distillation and extractive distillation have attracted attention for their capability to work with large flow rates [2]; however, they are high-energy consumption technologies. In this regard, process intensification can play a significant role. We understand intensification as any process modification achieving higher efficiency, lower expenses, more environmentally friendly operation, size reduction, or any combination of the above. Examples of process intensification in distillation are membrane distillation [3], HiGee distillation [4], cyclic distillation [5], dividing wall column (DWC) [6–9], and dividing wall extractive distillation [1,10–12], among others. In spite of the potential savings, intensified separation systems still represent a minor proportion on distillation sequences due to a more challenging control know-how [13].

Control property analysis by using condition number and minimum singular values, and dynamic responses studies have shown that intensified separation systems, including DWC and Petlyuk systems, can outperform conventional column systems [14–17].

In a previous work, through systematic process synthesis and intensification using thermal couplings and column section recombination, we generated different intensified separation systems for the lignocellulosic bioethanol separation problem [1]. A column section stands for a set of trays or packing where no external mass or heat transfer takes place [18]. The selected intensified systems presented comparable total annual cost (TAC) savings with respect to a reference

Abbreviations: σ^*, Maximum singular values; σ_*, Minimum singular value; AC, Absorption Column; CF, Centrifuge Filter; CSD, Control Structure Design; DAP, Diammonium Phosphate; DC, Distillation Column; DWC, Divided Wall Column; F, Flash; HMF, Hydroxymethylfurfural; IAE, Integral of the Absolute Error; *L*, Reflux Flowrate; NREL, National Renewable Energy Laboratory; NRTL, Non-Random Two-Liquid Model; PI, Proportional-Integral; SC, Stripping Column; SVD, Singular Value Decomposition; TAC, Total Annual Cost [USD year^{-1}]; TUC, Total Utility Coss [USD year^{-1}]; U, Direction of the process outputs; V, Direction of the process inputs; V, vapor boilup rate; wt.%, Mass Percentage; σ, Singular Values; Σ, Diagonal Matrix; γ, Condition number

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system (using conventional distillation columns). However, they had different separation train and diameters sizes, number and mass flowrate of recycles and total utilities costs (TUC) savings. Understanding how this intensification procedure (thermal couplings and column section recombination) modified these process features and their effect on control properties and dynamic responses can give further insights about which process features have more effect on control properties, and therefore suggest where the intensification should focus on. Moreover, it may also contribute to accelerate the selection from among intensified separation systems by using control shortcuts and analyzing process features in a relatively straightforward way.

Contrary to the conventional distillation process, control properties of intensified distillation columns have been little explored in the published literature, although some authors have attacked this problem. Jimenez et al. [19] have demonstrated the application of the singular value decomposition (SVD) technique to compare the controllability properties of intensified distillation structures. It is important to highlight that the dynamic model used in each equilibrium stage, for application of SVD, includes transient total mass balance, transient component mass balances, equilibrium relationship, summation constraints and transient energy balance. Similar control studies [20–22] have performed control analysis in studies of complex distillation systems. As far as we know, no study has been reported on the control properties in highly intensified distillation systems in the production of biofuels.

In the present work, we evaluated the control properties using SVD and dynamic responses (mild disturbances and set point changes) of different separation systems, as well as the effect of using thermal couplings and column section recombination as intensification tools with respect to the process features: diameter sizes, TUC savings and number and mass flowrate of recycles. This control test do not consider this process stage, indeed, analysis of operating procedures such as startup and shutdown strategies, which are transient and discontinuous by nature, so it can be considered as a separate study [23].

We expect to obtain good control properties and dynamic responses for the intensified separation systems, identify promising separation systems, and relate key process features with control properties and dynamic responses for the lignocellulosic bioethanol separation problem.

First, we explained how we selected the intensified separations systems; then, we described the evaluation methodologies followed by the most relevant results, and finally, we concluded with our observations regarding the relation between process features, and control properties and dynamic responses for the present case study.

2. Synthesis of new intensified separation systems for lignocellulosic bioethanol separation and dehydration

2.1. Separation problem and reference separation system

The separation stream of this work consisted in a mixture of gas (4.78 wt%), water and bioethanol (79.17 wt%) and soluble organic compounds (16.15 wt%). This mixture is the solids-free fermentation broth presented in previous works [1,2]. The composition and flow rate of the separation stream is described in Table 1. The reference separation system [1], is depicted in Fig. 1.

Shortly in Fig. 1, the solids-free lignocellulosic bioethanol stream was fed to the distillation column (DC-1) where most of the water and organic matter, in the way of stillage, were separated as bottom product, and the top stream sent to a set of two flashes (F-2 and F-3). F-2 and F-3 operated at different conditions and separated the gases producing a hydrous bioethanol stream sent to an extractive distillation column EDC-5. An absorption column (AC-4) recovered bioethanol dragged with the gases and sent it back to DC-1. Biothanol purity specification was achieved when glycerol and the hydrous bioethanol were fed in EDC-5. Finally, the recovery of glycerol was done by a combination of a flash (F-6) and a stripping column (SC-7). Finally, the recovered solvent can be recycled back to EDC-5.

Table 1
Mass composition (wt%) of the lignocellulosic bioethanol separation problem.

Lignocellulosic bioethanol (solids-free)		Grouped-components	
NH_3	0.01%	**Main gas components**	4.68%
O_2	0.01%		
CO_2	4.67%		
Bioethanol H_2O	4.89% 74.28%	**Bioethanol + water**	79.17%
Glucose	0.67%	**Soluble organic components**	16.15%
Xylose	0.61%		
Extractive	1.68%		
Soluble Lignin	0.33%		
HMF	0.24%		
Furfural	0.02%		
Lactic Acid	0.15%		
Xylitol	0.05%		
Glycerol	0.01%		
Succinic Acid	0.02%		
$(NH_4)_2SO_4$	11.76%		
NH_4 acetate	0.55%		
DAP	0.07%		
Total mass flow rate:		421,064 kg/h	

2.2. Design and simulation of the separation systems

We used the process simulator Aspen Plus V8.8, thermodynamic package NRTL, Henry gaseous components, NREL physical property data (components not included in Aspen properties database) [24] and RadFrac modules to simulate the separation systems. Design parameters and operating conditions were taken from Torres-Ortega and Rong [1].

We evaluated total annual cost (TAC) according to the modular methodology of Guthrie [25,26] using the simplified expression depicted in Eq. (1), considering five years of return of investment. We defined the total utilities cost (TUC) as the summation of each equipment utilities cost, Eq. (2).

$$TAC = \sum \left[\left(\frac{Capital\ Cost_i}{Time\ of\ investment} \right) + Utility\ cos\ ts \right] \tag{1}$$

$$TUC = \Sigma[\,Cos\,t\ Re\,b.\ duty_i + Cos\,t\ Cond.\ duty_i + Cos\,t\ Pump\,\&\,Comp\ power_i + Cos\,t\ Solvent_i] \tag{2}$$

We approximated DWC and other intensified systems modeling by using column sections system model, Fig. 2. This model reflects better the actual situation and allows for maximum flexibility regarding specifications, and vapor and liquid splits for control studies [6]. Equivalent approximated models have been experimental validated in several studies [27–30].

2.3. Intensification procedure for separation systems

We used thermal couplings and column section recombination as major intensification tools due to the possibility to have a sequential (synthesis and design) procedure that simplifies the whole task. That is, we start with a “conventional” separation system using conventional columns and designs, and then we can synthesize further intensified separation systems based on the previous conventional system. The details of the general procedure are thoroughly discussed somewhere else [1,31–36], to name a few.

The summary of the reference and intensified separation systems results of the work presented by Torres-Ortega and Rong [1] are depicted in Fig. 3. Briefly, process intensification was applied in the separation section (to obtain hydrous bioethanol) –in blue-, dehydration section (to obtain final product) –in green-and both separation and

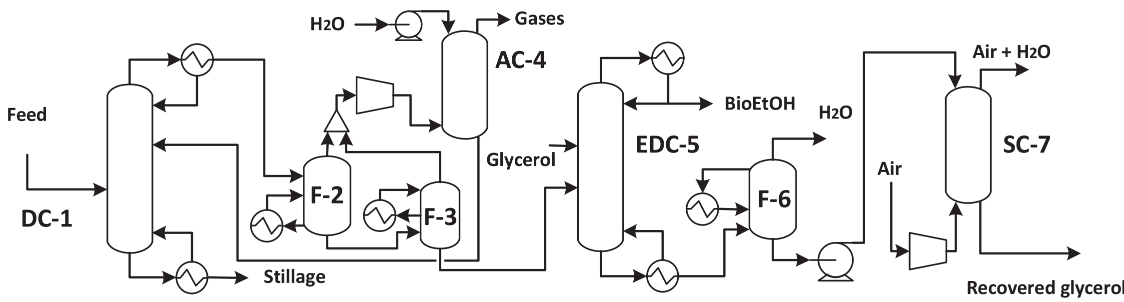

Fig. 1. Reference sequence (Ref_Seq) for the separation and dehydration of lignocellulosic bioethanol. DC, EDC, F, AC and SC stand for distillation column, extractive distillation column, flash, absorption column and stripping column.

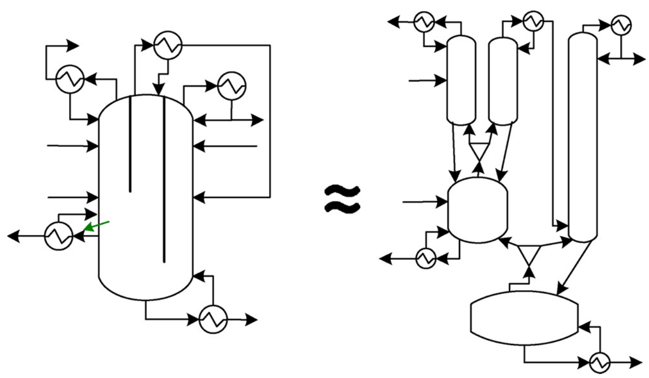

Fig. 2. Example of column section model implemented in Aspen Plus V8.8 for DWC_5 system.

dehydration sections simultaneously –in red.

Based on Fig. 3, it can be observed that most of the intensified systems met the design specifications and achieved relevant and comparable TAC savings with respect to the reference system.

Besides the reference system, three intensified systems, in thicker frames in Fig. 3, were selected to compare control properties and dynamic responses. First, Intens_3 represented the option with highest TAC saving in the separation-section intensification block, besides having the fewest amount of recycles. Second, DWC_3 presented one of the highest TAC and TUC savings, while including few recycles and in the dehydration-section intensification block. Finally, in the total-section intensification block, DWC_5 had the highest TAC and TUC savings, and the fewest number of recycles.

The intensification changes applied in the selected separation systems above are described in Fig. 4a) to c) for further discussion purposes.

3. Control properties and dynamic responses analysis for the lignocellulosic bioethanol separation and dehydration problem

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 reference sequence. We carried out two sets of analysis: (i) control properties of distillation by using singular value decomposition technique (SVD), and (ii) feedback dynamic response induced by mild set-point changes in product composition and flow rate disturbances.

3.1. Singular value decomposition: control properties

The SVD technique is used in order to estimate the natural dynamic properties of a system. Numerous works show this technique as a good way to define the dynamics of a system, even with this kind of DWCs

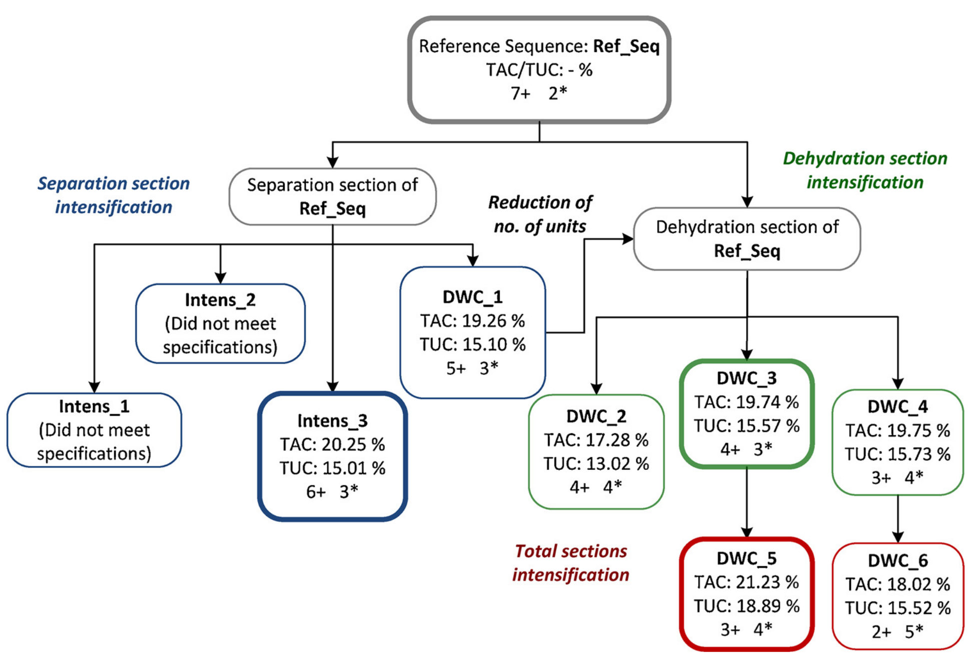

Fig. 3. TAC (total annual costs) and TUC (total utilities cost) percentage savings with respect to the reference system. The symbols "+" and "*"stand for the number of separation units of the system (separation train size), and the number of recycles, respectively.

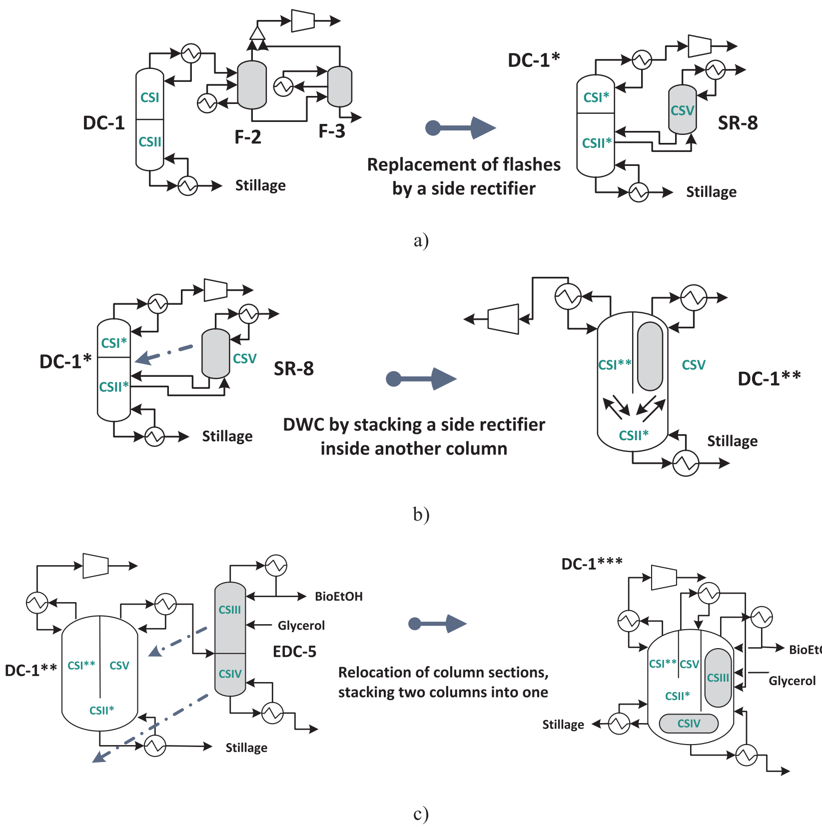

Fig. 4. Separation systems: a) Intensification change to generate Intens_3, b) Intensification change to generate DWC_3, c) Intensification change to generate DWC_5.
Notice: DC, F, AC, SC, SR, BioEtOH and CS stand for distillation column, flash, absorption column, stripping column, side rectifier, bioethanol and column section, respectively. Symbol "*" represents a change of a column section or a unit operation, i.e. CSI* represents CSI after one change by for example, adjusting the no. of trays; and DC-1** represents DC-1 after two changes by for example, multiple stacking processes.

with single and double diving wall [37–49]. Foremost, the steady-state simulations of the distillation sequences are exported to Aspen Dynamics as flow-driven simulations. In the dynamic situation, perturbations are imposed to the manipulated variables, by 0.5% over their nominal value. The responses of the measured variables are registered until a new steady state is achieved. For the open-loop analysis, purities of the key components have been chosen as reference.

After that, the dynamic responses are adjusted to transfer functions and ordered in a matrix. Then, by using a MATLAB routine, SVD is applied to each matrix of transfer functions. The mathematical expression of this matrix is represented by Eq. (3), which stands for the relative gain matrix of a linear system.

$$G = U\sum V^T \tag{3}$$

The diagonal matrix Σ contains the singular values σ_i ordered from largest to smallest, and two matrices U and V that are orthonormal. In practical terms, when G represents a process system, each singular value σ_i represents a mode i of operating the process and, according to this, the largest singular values indicate the most "energetic" modes. This interpretation generates the different control structure design (CSD) method that uses SVD as a tool. The CSD criteria are based on the maximum singular value, the condition number and the singular vectors:

- Maximum singular value (σ^*): It is desirable that the maximum singular value be small. In Havre et al.[35] this index was used as a criterion for selecting secondary measurements, a SVD analysis of the transfer functions that relate the output error with the disturbance and the uncertainty. The idea was to maintain this value small for all the frequency range of interest.
- Minimum singular value (σ_*): Morari [41] argues that this value should be big in order for a plant to have a good tracking and regulation performance, in case of limitations in the magnitude of inputs. By maintaining this value big, independent control of the variables can be guaranteed.
- Condition number (γ): This index is the ratio between the maximum and minimum singular values. The higher this value, the more difficult is the process control. A very high γ indicates that the plant tends to operate at certain modes and thus, the other modes would be difficult to attain. For this reason, a set that gives a system with a small γ should be selected.

In summary, low values of σ^* and high values of σ_* are desired so that the system can assimilate the disturbances. Therefore, small values of the condition number γ are desirable over large values [42]. However it must be considered this work as an analysis of the dynamic behavior of this process. In brief, open-loop test is a qualitative test that only provide information about any inconvenient to satisfy the entire set of control objectives (notwithstanding the control strategy to be used).

In this work, we considered three controlled variables based on product mass compositions: water composition in the stillage stream, CO_2 composition in the gases stream and bioethanol in the BioEtOH stream. The manipulated variables associated to the controlled variables were reflux ratios for CO_2 in the gases stream, and bioethanol compositions in the bioethanol stream, respectively; and reboiler's heat duty for water composition in the stillage stream. The streams containing the controlled variables as well as the equipment directly associated to the manipulated variables are highlighted in blue in Fig. 5.a)–d). Several techniques, such the relative gain array method, can be used to fix manipulated variables and controlled variable for a control study. In the case of distillation columns, however, such loops are fairly well established and used successfully in practice, at least for conventional columns. A well-known selection 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 as manipulated variables in the distillate and bottom outputs compositions [43,44].

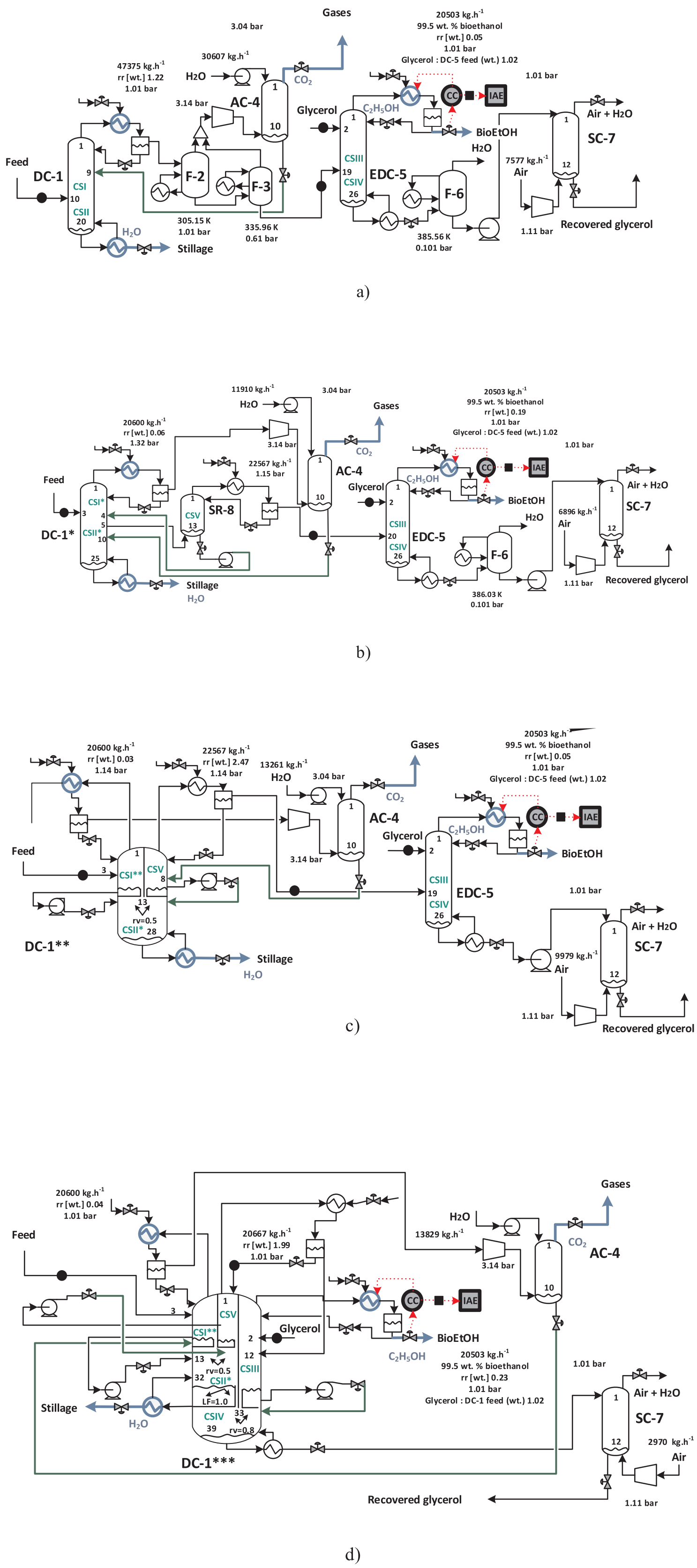

Fig. 5. Separation systems: a) Ref_Seq, b) Intens_3, c) DWC_3 and d) DWC_5. The streams and equipment highlighted in blue represent the elements used for the SVD analysis. The elements highlighted in gray represent the PI controllers set for the dynamic responses in Aspen Dynamics V8.8. The PI controllers associated to the bioethanol mass purity are in thick frames. The symbol ■ depicts where the set point change was applied; meanwhile the symbol ● depicts where the flow rate disturbances were applied. CS, DC, EDC, F, AC, SC, SR, rv, LF and BioEtOH stand for column section, distillation column, extractive distillation column, flash, absorption column, stripping column, side rectifier, ratio of vapor split, liquid flowrate split and bioethanol, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

In general, for feedback control is necessary a model that describes the effect of the inputs (flows) on the outputs (product composition). This does not implicate that the LV control structure is the preferred selection for control tests, the choice is made because L and V have a direct influence on composition and their effect is consequently only weakly dependent on the tuning of the level loops. This make also the most natural to consider the column model in terms of L and V as manipulated inputs [45].

3.2. Dynamic response: simple LV control structure for the bioethanol mass fraction

To supplement the SVD analysis, we carried out simulations in Aspen Dynamics V.8.8. We chose proportional-integral (PI) controllers, highlighted in gray Fig. 5.a)–d), based on their ample use for distillation systems in industrial practice [46–48]. First, we controlled sump levels by manipulating exiting liquid streams, controlled operating pressures by manipulating either exiting gas streams or condenser duty for distillation columns, and controlled drum levels in the distillation columns by manipulating overhead flowrates. With respect to DWCs, we controlled the sump levels of the sections next to the vertical wall(s) by manipulating the liquid streams leaving these sections. We assumed that the vapor streams, next to vertical walls, were kept in design range thanks to the controlled sump levels. For the control-loops above, we used the default-process simulator PI parameters. Second, we chose a LV control structure based on energy balances to control the bioethanol mass purity [49]. Therefore, the bioethanol mass composition in the product stream, which is obtained as an overhead product, was controlled by the reflux flowrate of the bioethanol total condenser in all the separation systems, highlighted in gray with thick frames in Fig. 5.a)–d). This type of control loops has been satisfactorily used in previous studies [43,50,51]. We tuned the PI parameters of the bioethanol mass purity controllers. The tuning procedure involved the reduction of the integral absolute error (IAE) for each loop for each distillation system [52]. The IAE reduction consisted in setting an initial value of the proportional gain, K_c; followed by a search over the integral reset time values, τ_i, until finding a local minimum value of IAE. Then, we repeated this process for several values of proportional gain until we did not achieve further improvement in IAE values. By doing this, we assumed there are not-unknown disturbances, as well as achievable plant stability during each tuning.

We studied the dynamic response of the controller associated to the bioethanol mass fraction for the reference and intensified distillation systems, Fig. 5.a)–d). The different cases were:

1.- Change on set point composition (−1 wt% bioethanol) for the BioEtOH stream.
2.- Flow rate disturbance (−1 wt% bioethanol) in the main feed stream.
3.- Flow rate disturbance (−5 and +5 wt% water) in the hydrous bioethanol stream.
4.- Flow rate disturbance (−5 and +5 wt% glycerol) in the glycerol stream.

4. Control properties and dynamic responses of the separation systems

4.1. SVD analysis results

At this point it is important to highlight that the dynamic open-loop responses used to generate the transfer functions were obtained considering the complete rigorous model in the transient state (including the hydraulic model), therefore the high non-linearity of the system is considered.

The control properties results are depicted in Figs. 6 and 7. At low frequencies (small disturbances), the systems Intens_3, Ref_Seq and then DWC_5 presented the highest minimum singular values and lowest condition numbers; however, the results from Ref_Seq were the most unstable along the range studied. A point to emphasize is that the results at low frequency (near the nominal value of the operation of the distillation columns) are the most representative of the dynamic behavior in the industrial operation [53] in comparison with results at high frequencies (high disturbances). However, even under these conditions, the systems are still feasible to be controlled but with a greater control effort. Therefore, we expect that Intens_3 and DWC_5 systems will exhibit better and more stable control properties under feedback control and better conditioning to mild disturbances than the other distillation systems.

In general, the results from the SVD analysis indicated that the intensified separation systems presented competitive control properties with respect to the conventional separation system under mild disturbances. These results are consistent with those reported, for example by Sánchez-Ramírez et al. [53], Segovia-Hernandez et al. [20] and Torres-Ortega et al. [54]. Moreover, note for both Figs. 6 and 7, the open-loop testing results are showed at low frequency, this it would represent a disturbance that allow the process to remain near the nominal state. In other words, to handle with small disturbances improve the representation of real process where common operation is near steady state. Besides, this small disturbances are theoretically represented by both condition number and minimum singular value at low frequency [55].

4.2. Simple LV control-structure dynamic response results

The PI parameters and the IAE values for the bioethanol mass purity are depicted in Table 2. Moreover, in Table 2, we summarized the PI parameters resulting from the tuning process and IAE values for the controller associated with the bioethanol mass purity. The separation system that presented the lowest IAE values for each dynamic case study was highlighted in grey.

Regarding the set point changes and disturbances on the controller associated to the bioethanol mass purity, we observed the follow:

- For the first case with change of set point (−1 wt% bioethanol) in the BioEtOH stream, Intens_3 and then DWC_5 systems presented the lowest values of IAE, (fastest dynamic responses). These results are consistent with those observed in Fig. 8 where Intens_3 and DWC_5 showed the lowest response times to reach the new steady state (less than 0.4 h). The reference system (Ref Seq) showed the longest time of stabilization and high deviation before reaching the new set point.
- For she second case, flow rate disturbance (-1 wt% bioethanol) in the main feed, the system DWC_5 and then Intens_3 presented the lowest IAE values, and shortest and most stable response times (lower than 1 h), Fig. 9. Again, Ref Seq presented the slowest dynamic responses.
- For the third case, flow rate disturbance (-5 and +5 wt% water) in the hydrous bioethanol stream, systems Intens_3 followed by DWC_5 presented the lowest IAE values and shortest and most stable response times (lower than 1 h); Figs. 10 and 11; meanwhile Ref Seq was the least favored system in both −5 and + 5 wt% cases.
- Finally for the fourth case, flow rate disturbance (−5 and + 5 wt% glycerol) in the glycerol stream. Again, Intens_3 followed by DWC_5 presented the lowest IAE values and fastest dynamic responses (lower than 1.5 h), Figs. 12 and 13, but this time DWC_3 was the system with the slowest response in both −5 and + 5 wt% cases.

In general, these results were consistent with those obtained by the SVD analysis (Figs. 6 and 7), confirming the intensified separation systems had competitive control properties and dynamic responses in comparison to the reference system for the lignocellulosic bioethanol separation and dehydration problem.

5. Process features modified during intensification and their effect on control properties and dynamic responses

Intensifying the separation system does not necessarily compromises the control properties and dynamic responses of the systems. In fact, for the present case study, overall the intensified systems presented competitive control properties and faster dynamic responses than the reference system (convention distillation columns).

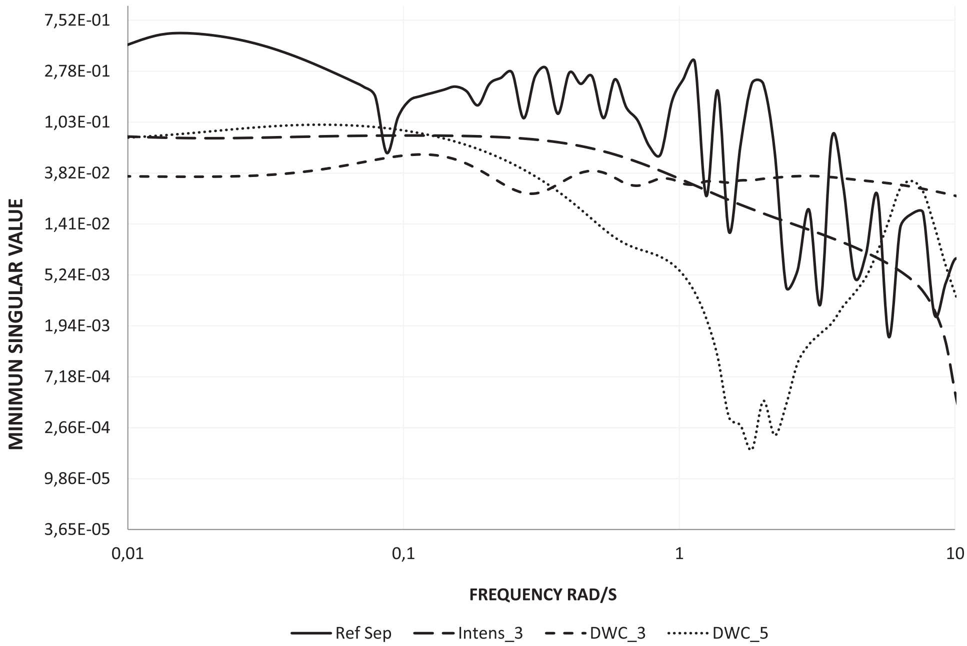

Fig. 6. Minimum singular value under different frequencies for the separation sequences.

Intensifying a separation sequence usually implies size reduction; and as a consequence, the dynamic response tends to be faster than a conventional separation system [13]. However, the intensification involves more sump and drum levels, pressure loops, more recycles, etc.

Moreover, there was a difference of performance among the intensified systems. This makes evident the interaction of different process features influencing control properties and dynamic responses. Therefore, we analyzed diameter sizes, number and liquid mass flowrate of recycles (green arrows in Fig. 5.a)–d)) and TUC savings of the systems studied. To enrich the discussion, we analyzed column sections (green labels in Fig. 5 a)–d)) instead of whole columns where relevant. Tables 3–5 present this information.

The first analysis corresponds to the performance comparison between the reference system and the intensified systems. When comparing the diameter sizes as in Table 3, we observed an important diameter size reduction in all intensified systems for the stillage separation, CSII, that representing more than 85 wt% in the feed stream, followed by another decrease in CSI (second column section with a high feed flow rate), separating gases from bioethanol and some water. Moreover, all the intensified separation systems presented a smaller AC-4 diameter as in Table 3, as well as smaller liquid mass recycles from AC-4 as in Table 4, back to DC-1. Regarding the extra recycles due to the presence of side rectifiers or vertical walls in the intensified systems, their mass ratio with respect to the main feed stream is lower than 0.06, not having a significant effect on their control properties or dynamic responses with respect to the reference system [56]. Finally, regarding utility costs as in Table 5, we can observe that the main reduction was in the heating costs, mainly associated to DC-1. When reducing DC-1 size and internal flowrates, we reduced the energy used and sped up dynamic responses. For the mild changes of set points and disturbances, the TUC savings did not represent dynamic restrictions. However, major changes of set points and disturbances need to be evaluated to analysis degree of nonlinearity and interaction, as well as loss of control degrees of freedom during operation [13,57].

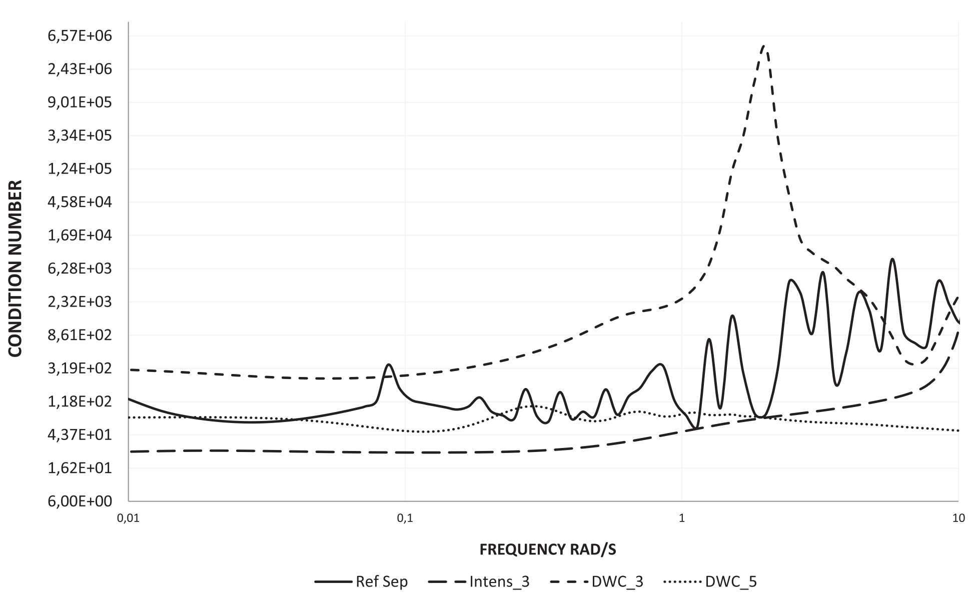

Fig. 7. Condition numbers under different frequencies for the separation sequences.

Table 2
Results of PI controllers tuning for the bioethanol mass purity stream.

Dynamic case	System	Kc	τi	IAE value
Composition Set-point −1 wt% Bioethanol	Ref_Seq	250	1	4.08E + 02
	Intens_3	250	1	7.93E + 01
	DWC_3	250	1	1.49E + 02
	DWC_5	250	3	9.01E + 01
Flow rate disturbance (−1 wt%) Bioethanol	Ref_Seq	250	20	1.11E − 03
	Intens_3	250	1	1.54E − 05
	DWC_3	250	15	1.60E − 04
	DWC_5	250	3	1.18E − 05
Flow rate disturbance (−5 wt%) Water	Ref_Seq	250	20	2.32E − 04
	Intens_3	250	20	6.94E − 06
	DWC_3	250	20	6.17E − 05
	DWC_5	250	3	8.97E − 06
Flow rate disturbance (+5 wt%) Water	Ref_Seq	250	20	2.32E − 04
	Intens_3	250	20	6.94E − 06
	DWC_3	250	20	6.17E − 05
	DWC_5	250	3	8.97E − 06
Flow rate disturbance (−5 wt%) Glycerol	Ref_Seq	250	1	9.04E − 05
	Intens_3	250	20	4.10E − 05
	DWC_3	250	20	2.33E − 04
	DWC_5	250	3	4.25E − 05
Flow rate disturbance (+5 wt%) Glycerol	Ref_Seq	250	1	1.19E − 04
	Intens_3	250	20	3.80E − 05
	DWC_3	250	20	2.42E − 04
	DWC_5	250	3	5.01E − 05

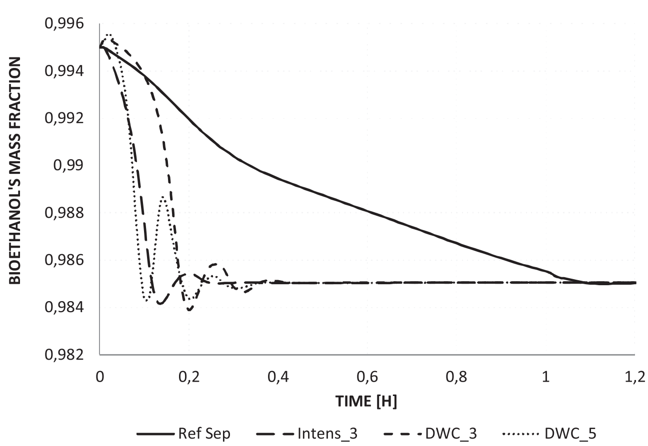

Fig. 8. Dynamic responses for a change of set point (−1 wt% bioethanol).

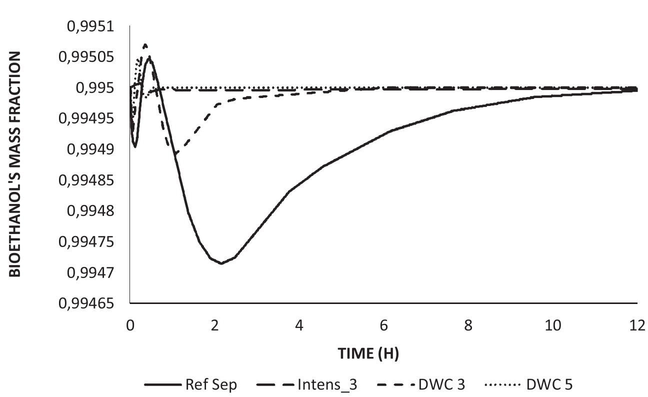

Fig. 9. Dynamic responses for flow rate disturbance of bioethanol in the main stream (−1 wt%).

Second analysis compares the performance between intensified systems. Intens_1 has the lowest diameter size for CSII (stillage separation) as in Table 3, as well as the associated AC-4 diameter and respective recycle as in Table 4, representing the major contribution. Besides this, it has the lowest diameter for concentrating the bioethanol

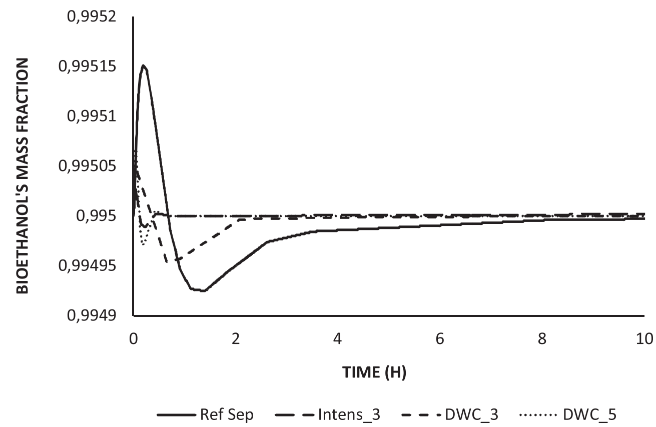

Fig. 10. Dynamic responses for flow rate disturbance of water in the hydrated bioethanol stream (−5 wt%).

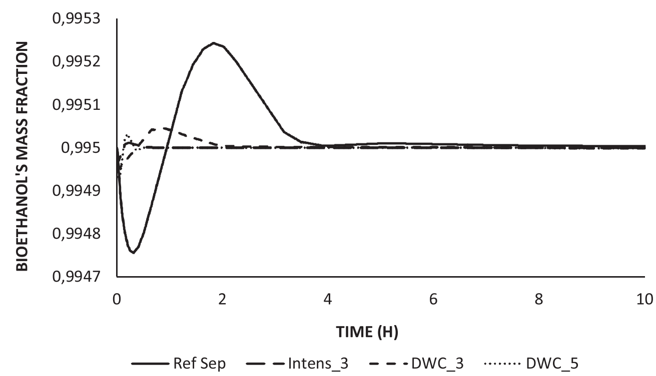

Fig. 11. Dynamic responses for flow rate disturbance of water content in the hydrated bioethanol stream (+5 wt%).

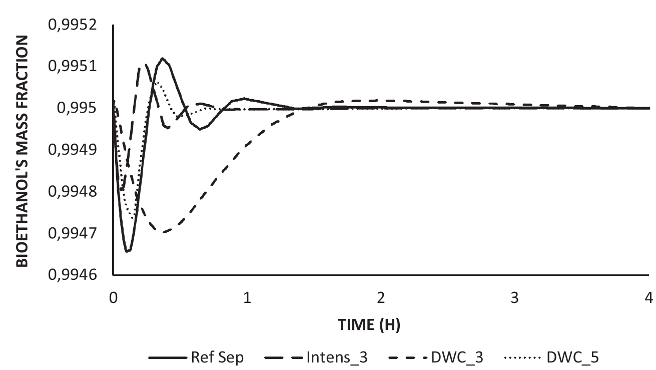

Fig. 12. Dynamic responses for flow rate disturbance of glycerol in the dehydration feed (−5 wt%).

(90 wt%) and a relatively low mass recycle for SR-8. These features, besides the fact that fewer level and pressure controls are required, make this system the most competitive for further studies. After Intens_1, DWC_5 showed faster dynamic responses than DWC_3. Regarding CSII diameter (stillage separation) DWC_5 size diameter was 0.78% higher than DWC_3, neglecting the influence in the dynamic responses. Regarding CSI, CSIII and CSV diameter sizes, DWC_5 presented diameter sizes slightly bigger by 5.3% or lower with respect to DWC_3. We considered the significant process features giving DWC_5 faster dynamic responses than DWC_3 are the CSIV diameter (glycerol separation) and mass recycle in CSV (concentration of bioethanol to 90 wt%). CSIV diameter and liquid mass recycle in CSV are 90% and 22.24% smaller in DWC_5 than DWC_3.

Finally, we present in Fig. 14 our major findings when using thermal

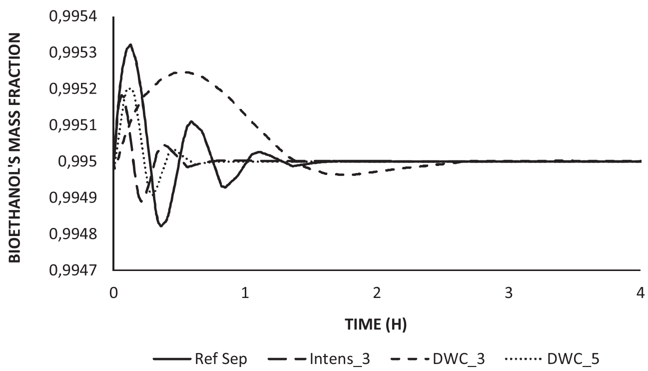

Fig. 13. Dynamic responses for flow rate disturbance of glycerol in the dehydration feed (+5 wt%).

Finally, we reduced the set of intensified separation systems with better control properties to two systems: first and simplest design and construction Intens_3, followed by DWC_5.

couplings and column section recombination as intensification tools and their effect on process features and control properties of the lignocellulosic bioethanol separation system. Before applying these intensification tools, identify the largest column section(s) and their mass flow rate recycles, as they may contribute the most for energy consumption and delays in dynamic responses. Then, track how these process features change through the individual intensification changes, and prioritize those changes than lead to smaller column sections and mass flow rate recycles.

6. Conclusions

In the present work, we had a reference separation system (conventional distillation columns) as in Fig. 1, and a set of intensified separation systems with similar cost savings with respect to the reference system, and we focused on studying how thermal couplings and column section recombination tools affected process features and control properties for these systems. We evaluated control properties by SVD (singular value decomposition) study and mild set point changes and disturbances using PI controllers and a LV control structure for the bioethanol mass purity. Overall, the intensified systems presented competitive control properties (higher minimum singular values and lower condition numbers), and faster dynamic responses (more quickly stabilization to new steady states) than the reference system, although they required a higher number of sump and drum level controllers, as well as pressure controllers. Among the intensified systems, Intes_3 presented the most suitable process feature changes for control property purposes.

Moreover, for the present case studied, insights regarding process feature changes and control properties during application of thermal couplings and column section recombination are obtained. Understanding these insights may speed up the process of selecting promising intensified separation systems. Briefly, process features changes leading to smaller key column section diameters and their respective mass flow rate recycles represent systems that besides saving energy, leads to better control properties and since the diameter reduction of the column and the reduction of internal flows is associated with decreasing the sizing of the column and diminution of energy consumption, therefore the total annual cost of the system is diminished. Thus, improving the control properties implicitly reduces the total annual cost of the configuration for this case study.

However, the procedures presented in this work do not compare at same design stage both the control properties and TAC evaluation. The complexity of that study remains in using parallel both models (steady-state and dynamic state) for early design decisions. In general terms, this methodology which evaluates control properties as subsequent study has generated a relative good understanding of dynamic properties of this kind of intensified systems [52,53]. Otherwise, it is necessary to make some reduction in the entire dynamic model, or even consider (in the case of SVD analysis) a frequency cero analysis to perform an equal weighting between cost and control properties [58].

However, further detailed control studies and plantwide analysis are still required to confirm and supplement the above insights.

In summary, the main contributions of this work are:

- Control property studies for the first time in these new lignocellulosic bioethanol separation systems.
- Understanding of how thermal couplings and column section recombination affects process features change and control properties, as well as identify specific process features changes positively influencing control properties.

Competing financial interest

The authors declare no competing financial interest.

Table 4
Liquid mass recycles for different column sections/columns for the separation systems.

	Recycle function	Recycle from SR-8/CSV ($kg\,h^{-1}$) BioEtOH (90% wt.)	Recycle from AC-4 ($kg\,h^{-1}$) BioEtOH recovery	Recycle from CSIII ($kg\,h^{-1}$) BioEtOH (99.5% wt.)
SYSTEM	**Ref_Seq**	–	34,208.10	–
	Intens_1	19,733.01	12,292.72	–
	DWC_3	21,581.36	14,290.30	–
	DWC_5	17,655.45	14,409.77	31,136.72

Table 5
Utility costs break down for the separation systems.

		Total utility cost (MUSD y^{-1})	Cooling cost (MUSD y^{-1})	Heating cost (MUSD y^{-1})	Electricity cost (MUSD y^{-1})	Solvent cost (MUSD y^{-1})
SYSTEM	**Ref_Seq**	35.81	1.22	33.81	0.62	0.16
	Intens_1	30.43	0.29	29.74	0.26	0.15
	DWC_3	30.23	0.20	29.62	0.31	0.11
	DWC_5	29.04	0.35	28.16	0.33	0.21

Table 3
Diameter sizes for different column sections (CS) or columns for the separation systems. Note: SR-8 and AC-4 stand for side rectifier and absorption column, respectively.

	CS function	CSI (m) Gas separation	CSII (m) Stillage separation	SR-8/CSV (m) BioEtOH (90% wt.)	CSIII (m) BioEtOH (99.5% wt.)	CSIV (m) Glycerol separation	AC-4 (m) BioEtOH recovery
SYSTEM	**Ref_Seq**	5.27	5.27	–	1.68	1.68	1.35
	Intens_1	4.87	4.87	2.77	1.71	1.71	1.12
	DWC_3	3.8	5.1	2.88	1.71	1.71	1.14
	DWC_5	3.88	5.14	3	1.8	0.9	1.2

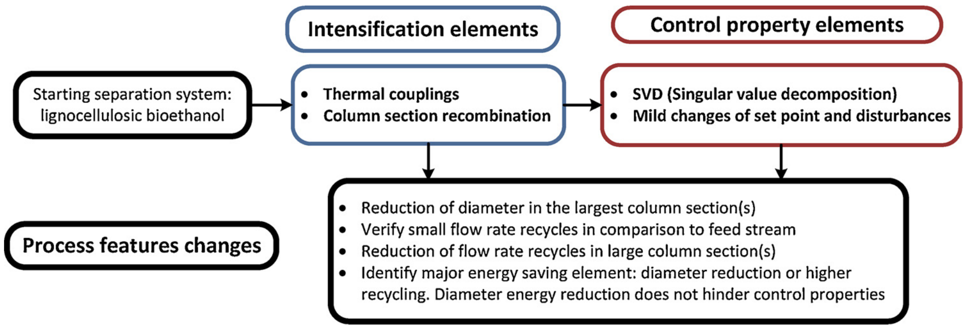

Fig. 14. Process features changes during thermal coupling and column section recombination that lead to separation systems with potential competitive control properties.

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