

Analysis of Control Properties as a Sustainability Indicator in Intensified Processes for Levulinic Acid Purification

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ABSTRACT

The evaluation of control properties in industrial processes is essential to achieve sustainability, a very relevant topic today. This study emphasizes the importance of control studies to ensure that processes are efficient, operable and safe. While strategies such as process intensification can reduce the size, cost, and consumption of energy, it can present challenges in control and operability. This work focuses on the evaluation of the control properties of schemes with different degrees of intensification for the purification of levulinic acid, with the aim of identifying designs with the best control properties and the best economic and environmental indicators. The schemes were designed under a systematic synthesis strategy and optimized using the hybrid method of differential evolution with a tabu list, considering the total annual cost and Eco-indicator 99. An open-loop study analyzed the relationship between manipulable variables and output variables using total condition number, sensitivity index, and relative gain matrix analysis. The dynamic behavior in a closed loop was subsequently analyzed using the minimization of the absolute error integral as a criterion. The results showed that the design, which includes a liquid-liquid extraction column, three distillation columns, and thermal coupling, presented the best dynamic performance. This design had a low total condition number, a below-average sensitivity index, a stable control structure, and low values of the absolute error integral. In addition, it stood out for its excellent cost and environmental impact indicators, which makes it the most favorable option among the proposed designs.

Keywords: Stochastic optimization, Control, Distillation, Bioproducts.

INTRODUCTION

The obtaining of bioproducts from lignocellulosic biomass as a sustainable alternative has gained strength in recent years. However, the processes for obtaining these bioproducts generate very dilute product streams, which represents a challenge due to the high energy consumption that their purification entails [1].

An alternative to guarantee the viability of obtaining bioproducts is the use of Process Intensification (PI). PI is an ideology that is born from the need to increase the efficiency of processes, saving energy, and maximizing production and profits, while at the same time reducing emissions of CO₂ and other by-products or polluting wastes [2]. The PI has been implemented in distillation,

obtaining hybrid schemes, such as reactive distillation that combines reaction and separation, as well as HiGee columns that have external assistance [3]. Furthermore, if non-reactive hybrid separation PI is considered, there are schemes such as dividing wall columns (DWC), Petlyuk columns and thermally coupled columns present significant energy savings, obtaining savings of more than 25%.

PI is a powerful strategy for creating more efficient, economical, safe, and environmentally friendly processes. However, the industrial operability of these intensified processes can be challenging due to their complexity and the need for sophisticated control structures [4]. Some studies show that intensified designs may have better control properties, while others suggest that

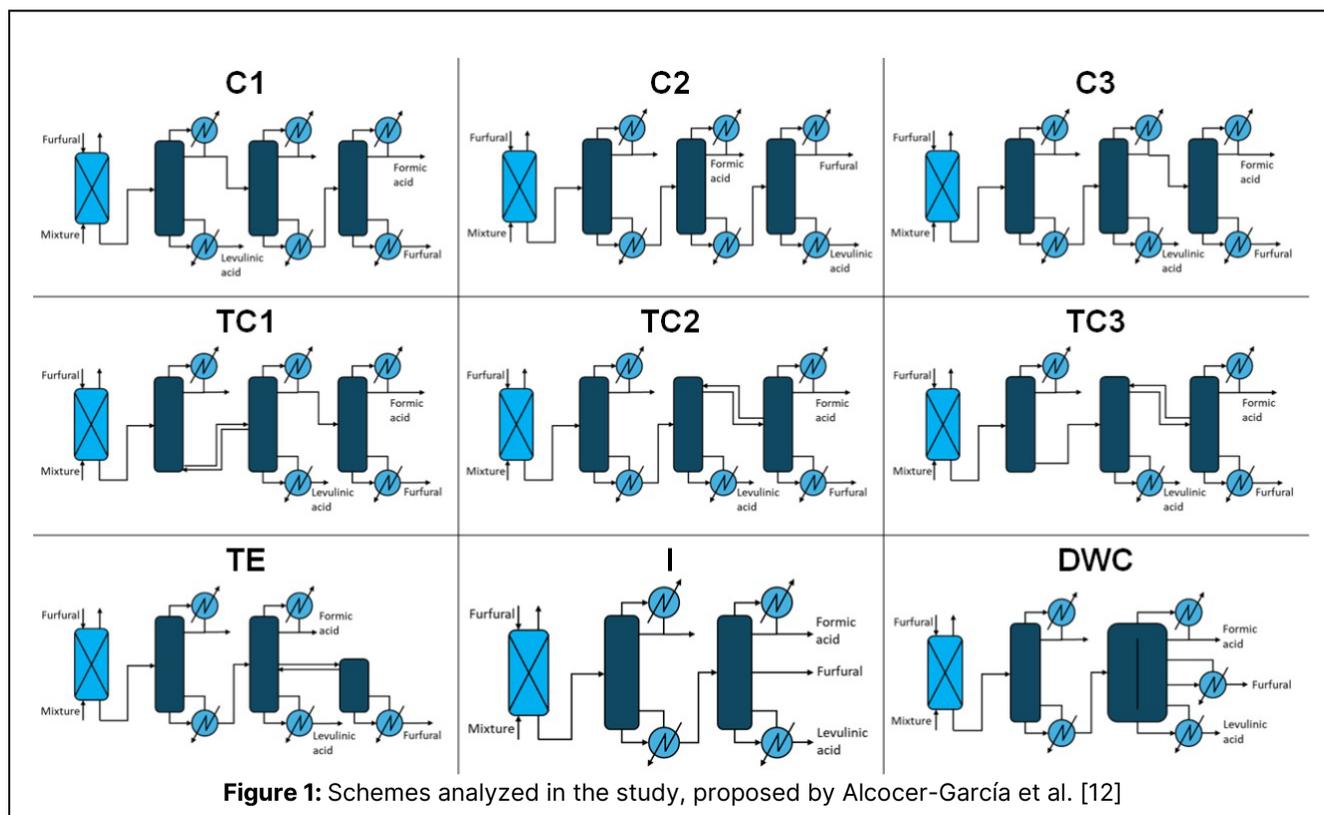


Figure 1: Schemes analyzed in the study, proposed by Alcocer-García et al. [12]

increased complexity does not always improve control. An in-depth analysis of control properties is essential to develop effective strategies and contribute to more sustainable and efficient processes. Specific challenges include modeling and parameter tuning, complex dynamic behavior, and reduced degrees of freedom. Addressing these challenges allows progress toward the Sustainable Development Goals (SDGs) and the transition to Industry 4.0, promoting more precise and sustainable production.

Incorporating process operability indices during design allows for the development of efficient, economically viable, and dynamically stable chemical processes. To analyze the control of distillation systems and other industrial systems, techniques and indices have been developed that help to understand their dynamic behavior. Molina-Guerrero et al. [5] performed an open-loop control analysis of an extractive distillation system, while Li et al. [6] focused on minimizing the IAE for systems with thermally coupled columns. Pushkala et al. [7] studied the optimal control structure for a reactive distillation column using the RGA. Alcocer-García et al. [8] found that schemes with greater intensification do not always have the best control properties. Therefore, dynamic analyses are essential to understand the behavior of processes and build effective control strategies.

On the other hand, levulinic acid (LA), considered one of the twelve main bioproducts through biomass, presents a bottleneck in its separation process. This is because the current resulting from thermal pretreatment

and acid hydrolysis generates a very dilute current, causing the formation of azeotropes and increasing the cost of their separation, and consequently the production cost [9]. Recently, Alcocer et al. [10] developed schemes with varying degrees of intensification for the purification of levulinic acid, demonstrating significant economic and environmental savings. However, these schemes lack a comprehensive analysis of control properties. Consequently, this study undertakes an in-depth analysis of the control properties for the systems proposed by Alcocer et al. [10], thereby contributing to the understanding and enhancement of operability in intensified systems and to obtain safer and more sustainable designs.

The schemes studied consist of conventional sequences (C1, C2, C3), thermally coupled (TA1, TA2, TA3), thermally equivalent (TE), intensified with $n-1$ columns (I) and with dividing wall (DWC) were obtained, all with a previous liquid-liquid extraction process, shown in Figure 1. These schemes were optimized using the hybrid method of differential evolution with a tabu list, taking as a multi-objective function the total annual cost (TAC) Eco-Indicator 99 (EI99). The analysis of the control properties was carried out using two approaches: open loop and closed loop. In the open-loop analysis, the indices of total condition number, RGA and power sensitivity were incorporated as indicators, as well as a closed-loop analysis seeking to minimize the IAE by tuning the gain and integral time of the controllers. The objective is to determine the dynamic properties of the schemes to predict

and resolve control problems. The collected information on TAC, environmental impact using the E199, and control properties provides a comprehensive understanding of the schemes and their industrial viability.

METODOLOGY

An open-loop analysis was performed using three indicators: γ , which measures directionality in multiple-input, multiple-output (MIMO) systems; the RGA, which evaluates the interactions between the output variables and the manipulable variables; and the SI, which assesses the sensitivity of the system to disturbances in feeding. The schemas were simulated in Aspen Plus and exported to Aspen Dynamics to obtain dynamic responses to disturbances, using the NRTL-HO thermodynamic model. The NRTL-HOC thermodynamic model, suitable for highly non-ideal chemical systems, was employed to calculate liquid-vapor equilibrium and the thermodynamic properties of the vapor phase of organic acids.

For the open loop, 0.5% perturbations were applied to the manipulable variables to calculate the following indices: Total Condition Number (TCN), Relative Gain Matrix (RGA), and Sensitivity Index (SI). The responses obtained were fitted to a transfer function in MATLAB and transformed into the frequency domain to calculate γ and RGA. The open loop methodology is shown in Figure 2.

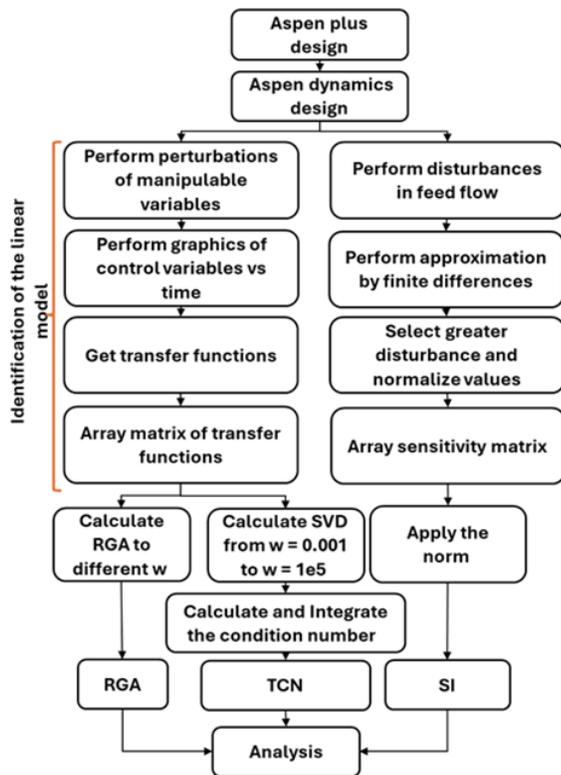


Figure 2. Open-loop methodology.

TCN will allow us to identify the input-output control relationship of the system. This will be calculated using the method of Santaella et al. [11], where the integral of the crow generated by the condition number γ and the frequency ω is calculated.

$$TCN = \int_{\omega_1}^{\omega_2} \gamma d\omega \quad (1)$$

A high γ suggests that the system is sensitive to input uncertainties, but it does not always imply greater sensitivity in practice. Systems with a lower γ are expected to have better dynamic performance and be easier to control, as they are less sensitive to disturbances. This facilitates the implementation of simple and straightforward control strategies, such as Proportional-Integral-Derivative control.

The RGA analysis shows how the manipulable variables affect the system's outputs. In distillation columns, the most used control structure is the LV configuration, which controls the distillate composition with the reflux ratio (RR) and the bottom composition with the reboiler heat duty (RHD). However, in intensified schemes, different structures can be identified at LV (DLV), where a different manipulable variable than the one established in the LV configuration has a greater influence on the output variable. In addition, it is possible that some configurations at different frequencies have a changing control structure (CCS) entre la LV y DLV. The RGA on the other hand will allow us to find the best control pairing, calculated as follows:

$$RGA(G) = \Lambda(G) \triangleq G \otimes (G^{-1})^T \quad (2)$$

In this study evaluated the consistency of the control structure by calculating the RGA at three frequencies: steady state (0 rad/h), near steady state (0.001 rad/h), and high frequency (1e5 rad/h). The variation in the control structure at different frequencies indicates frequency dependence, suggesting nonlinearity and variations in interactions between process variables, which can affect control effectiveness and require adaptive or robust control strategies.

The SI allows us to observe the sensitivity of the output variables according to the input perturbations, which was calculated using the method of Prado-Rubio et al. [12] through finite differences:

$$SI(t) \approx \frac{X[F_{in}(t) + \Delta F_{in}(t)] - X(F_{in})}{\Delta F_{in}} \frac{F_{in}(0)}{X(0)} \quad (3)$$

A high SI value suggests that the system is very sensitive to disturbances in the feed, which requires the implementation of robust control strategies. This can include predictive control to anticipate and correct disturbances, as well as adaptive control to adjust parameters in real-time and maintain process stability and efficiency.

For the closed loop, perturbations were performed at the output of the LA at 5% and 30%, varying and tuning

the gain (K_c) and the integral time (τ_i) to find the minimum IAE using PI proportional-integral composition controllers. This with the objective of exploring dynamic behavior to a small disturbance and another of greater magnitude.

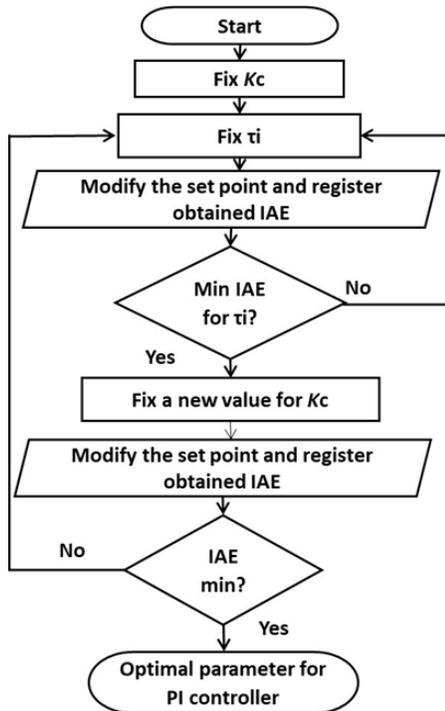


Figure 3. Process diagram for tuning PIC.

RESULTS

The open-loop analysis, shown in Table 1, revealed that the C2, TA1 and TA2 designs have an LV configuration at all frequencies, which translates into systems that can be controlled with direct and less complex strategies, reducing the need for advanced and expensive control systems. In particular, the TA2 design is distinguished by the lower TAC and IAE values, making it the most economical option with the lowest environmental impact. Therefore, considering that it presents a stable control structure, it is positioned as one of the best alternatives of the schemes studied.

On the other hand, the C3, TE, and DWC designs have a DLV configuration, which is stable, but may need more specific control. In these cases, a more specific control refers to the need to adjust and tune the manipulable variables more precisely to maintain the desired performance of the system. This could include the use of advanced control techniques such as predictive control, which anticipates future changes and adjusts control variables accordingly, or adaptive control, which adjusts controller parameters in real-time to respond to variations in process behavior. Although the TAC and EI99

values are slightly higher, they are still good alternatives. However, the C1, TA3 and TE designs, with CCS structure, are unstable and require advanced control strategies. Advanced control strategies are necessary for these designs because of the added complexity and nonlinear interaction between process variables. These strategies can include the use of nonlinear control, which handles the complex, nonlinear relationships between variables, and robust control, which is designed to maintain system performance under a wide range of uncertainty and disturbance conditions. It may also be necessary to implement multivariable control systems, which coordinate multiple inputs and outputs simultaneously to optimize the overall performance of the system.

Due to the increase in interconnection flows in the generated recycles, the TA3 design, which has two thermal couplings, has a greater sensitivity to disturbances in the power supply, generating an increase in SI. And also negatively impacting the TAC and HAI, being the design with the worst values in these indicators and presenting an unstable control structure, which positions it as the worst alternative of those studied.

Design factors such as column diameter and location of thermal couplings significantly impact TCN, with smaller diameter columns facilitating more effective control and reduced operating costs. In summary, systems with low TCN and SI values are more stable and less sensitive to disturbances, while systems with high values require more sophisticated and costly control strategies.

Table 1: Open-loop results analysis

Design	TAC	EI99	TCN	SI	RGA
C1	1.85E+07	5.35E+09	5.87E+09	13.37	CCS
C2	1.56E+07	4.70E+09	1.04E+07	6.68	LV
C3	1.49E+07	4.60E+09	1.95E+08	12.37	DLV
TA1	1.64E+07	4.56E+09	4.68E+09	0.50	LV
TA2	1.39E+07	4.21E+09	6.44E+06	11.28	LV
TA3	1.90E+07	5.16E+09	1.78E+07	33.15	CCS
TE	1.43E+07	4.35E+09	1.18E+07	11.50	DLV
I	1.77E+07	5.22E+09	6.30E+07	7.56	CCS
DWC	1.43E+07	4.34E+09	1.23E+07	11.49	DLV

The results of the closed-loop analysis are shown in Figure 4. Design TA1 presented the lowest IAE values in all perturbations due to the recycling between the first two distillation columns, which helps dampen disturbances. Additionally, this design shows the smallest K_c values, which improves stability and reduces oscillations. On the other hand, design C1 has the highest IAE values because it recovers levulinic acid in the first distillation column, complicating thermodynamic stability. Designs C2, C3, TA2, TA3, TE, I, and DWC stand out for having similar IAE values in all perturbations, revealing homogeneous dynamic behavior. This suggests that despite differences in configurations, their response to disturbances is uniformly balanced, facilitating a more

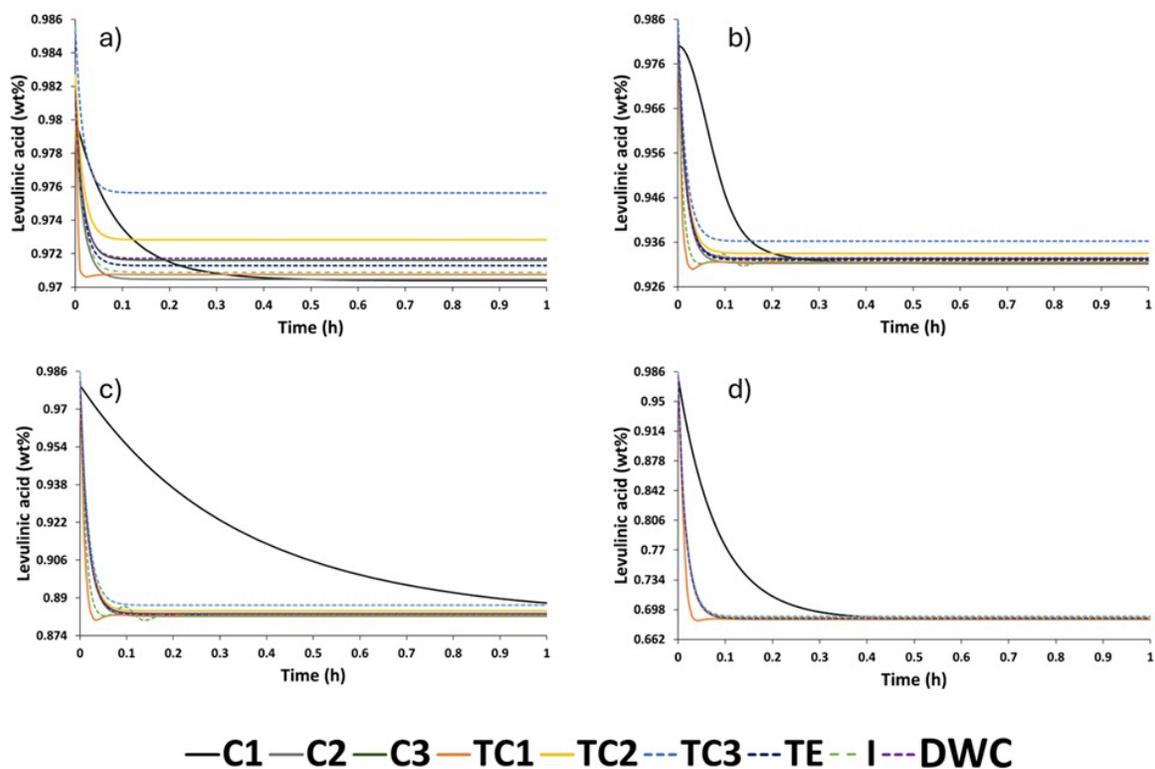


Figure 4: Closed-loop responses 1% (a), 5% (b), 10% (c) and 30% (d).

predictable implementation of control strategies.

These findings not only contribute to the understanding of operability in intensified systems but also highlight the importance of selecting appropriate configurations and optimizing specific factors to achieve effective control. The incorporation of additional thermal couplings, as in the TA3 design, improves the distribution of disturbances and the stability of the System in the face of disturbances in the manipulable variables, but makes it unstable in the face of disturbances in the feed.

In addition, sensitivity analysis and interactions between manipulable and output variables underscores the need for robust and adaptive control strategies, especially in designs with higher TCN and SI values. Finally, the economic and environmental benefits derived from the optimization and control of these systems underscore the relevance of these approaches to move towards a more sustainable and efficient industry. In particular, the TA2 design is positioned as the most balanced choice in terms of stability, control efficiency, and costs, which is crucial for the successful implementation of intensified processes in industrial practice. Design TA2 is low TAC and EI99 not only reflect its economic efficiency, but also its lower environmental footprint, standing out as a model for future sustainable designs in the industry.

CONCLUSIONS

The analyses conducted on open and closed loops revealed significant differences in the dynamic performance and control requirements of the various designs evaluated. Notably, the C2, TA1, and TA2 designs, which exhibit LV configurations at all frequencies, stand out for their superior dynamic performance and stability, obtaining the best values in RGA and IAE analyses and showing a homogeneous response to disturbances.

In contrast, designs such as C3, TE, and DWC, while stable in DLV configurations, may require more specific control techniques like predictive or adaptive control to maintain desired performance. Although the TAC and EI99 values are slightly higher, they remain good alternatives. However, C1, TA3, and TE designs with CCS structures proved unstable and necessitate advanced control strategies like nonlinear and robust control to handle complex interactions.

Factors such as column diameter and the location of thermal couplings significantly impact TCN, with smaller diameter columns facilitating more effective control and reduced operating costs. The TA2 design is particularly noteworthy, presenting the best TAC and EI99 values, making it the most economical option with the least environmental impact. The results underscore that systems with low TCN and SI values are more stable and less

sensitive to disturbances, allowing for more predictable and efficient control, while systems with high values require sophisticated control strategies.

The economic and sustainability benefits from optimizing and controlling these systems highlight their relevance to a more sustainable and efficient industry. The TA2 design stands out as the most balanced option in terms of stability, control efficiency, and costs, with its LV RGA configuration ensuring a stable and predictable control structure. These findings emphasize the importance of selecting suitable configurations and optimizing specific factors like column diameters and thermal couplings to achieve effective control, lower environmental impact, and reduced operational costs. This study provides a clear path toward optimizing the stability and efficiency of control systems in distillation column design.

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