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# Design and control of a distillation sequence for the purification of bioethanol obtained from sotol bagasse (Dasylirium sp.)



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# ABSTRACT

Sotol bagasse (SB) is a byproduct generated during the fermentation of the sotol plant (Dasylirium sp.) for the production of a traditional alcoholic beverage in Mexico. In this study, a novel ethanol purification distillation sequence is proposed, utilizing ethanol produced from SB treatment as described by González-Chavez et al. (2022). Prior to purification, the ethanol-water mixtures exhibited ratios of 0.054 and 0.935. The proposed distillation sequence comprises three columns: i) depletion, ii) extraction (employing ethylene glycol as the solvent agent), and iii) solvent recovery. To assess energy consumption and controllability, three solvent:feed ratios (1.5:1, 2:1, 2.5:1) were examined, taking into account the feed to the second column. The lowest energy consumption was observed at  $31 \times 10^3$  kW, corresponding to the 1.5:1 ratio. Optimal controllability was achieved at a solvent:feed ratio of 2.5:1, as evidenced by the lowest Integral Absolute Error (IAE) value of  $3.3 \times 10^{-3}$ and the highest proportional gain (Kc) value of 250. In summary, we leveraged previously reported experimental data to design a new distillation sequence with potential applications in the ethanol purification derived from SB fermentation. The outcomes of this investigation underscore the importance of promoting circular economy practices, particularly in the northern region of Mexico, where significant quantities of agrowastes are generated.

#### 1. Introduction

Regarding emerging industrial residual biomasses in Mexico, we find Sotol bagasse (SB) (Dasylirium sp.). SB is a plant biomass residue generated during the production of the alcoholic beverage known as "sotol". This plant is endemic to northern Mexico and is distributed in the states of Chihuahua, Coahuila, Sonora, and Zacatecas. Also, it can be found in southern United States (Texas, New México, and Arizona). Fermented sotol was part of Native Americans diet before the Spaniards introduced the process of distillation. To date, few options are available that consider SB for revaluation purposes (Flores-Gallegos et al., 2019). Recently, several researchers have determined the potential use of SB for biofuel production. For instance, González-Chavez et al. (2022) designed a biotechnological process based on a conventional biochemical platform (CBP) for the transformation of SB into second-generation

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bioethanol. Also, Piñón-Muñiz et al. (2023) used a biotechnological approach that included anoxic conditions for the transformation of SB into liquid biofuels. Through this process, these researchers obtained the mixture Acetone-Butanol-Ethanol (ABE).

However, to the best of our knowledge, the conceptual design of a complete SB biorefinery has not been reported. Previously, (González-Chavez et al., 2022) reported the design of the fundamental stages for the processing of this agro-residue based on a conventional biochemical platform (CBP); however, the design of the distillation stage was not reported. Herein, we consider that previous data obtained from other biomasses including wheat straw, apple bagasse, and sugarcane bagasse, among others, can be used to design new processes for the transformation of SB (López-Ortega et al., 2021; Molinuevo-Salces et al., 2020; Sanchez et al., 2013) The conceptual designs of biorefineries based on a CBP rely on four main stages: a) pretreatment; b) enzymatic

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hydrolysis; c) fermentation; and d) downstream process (Pino et al., 2018). Particularly, the downstream process is an essential step and it requires high amounts of energy. In the case of bioethanol, the literature reports sequences focused on the purification of bioethanol-water streams under different purification schemes (Ramírez-Márquez et al., 2013a; Vázquez-Ojeda et al., 2013). The use of columns in extractive distillation for ethanol dehydration results in high production capacities at industrial scales. Besides, it allows minimization of energy usage. For this reason, they are among the most studied processes of this type (Errico et al., 2015). In these strategies, a solvent agent with a high boiling point is used to increase the relative volatility of the component of interest (Ramírez-Márquez et al., 2013a). In summary, a wide variety of distillation sequences and solvent agents have been evaluated. They include simple columns and dividing-wall columns, among others (Oseguera-Villaseñor et al., 2018; Torres-Ortega et al., 2018). In every single case, the main objective is to reduce energy consumption and capital investment.

Errico et al. (2015) stated that in order to find a proper extractive distillation sequence for ethanol recovery from a specific azeotropic mixture, distillation sequences already reported in the literature could be used and adapted. Extractive distillation for bioethanol purification has been a focal point of extensive research, particularly in the realm of controllability studies, elucidating critical facets of system design and operation. (Ramírez-Márquez et al., 2013b) have been pivotal in open-loop investigations, not only optimizing system configuration but also identifying key parameters to augment the controllability of bioethanol extractive distillation. Closed-loop inquiries, spearheaded by (Gil et al., 2012), have delved into the dynamic analysis of the system, probing its response to external perturbations and scrutinizing its stability under feedback conditions. This line of research has found resonance in the work of (Arifin and Chien, 2008), who have explored advanced control strategies with a specific focus on enhancing controllability during bioethanol extractive distillation. (Hernández-Vargas et al., 2023) have significantly advanced the field by investigating the impact of specific variables on the efficiency of the extractive distillation process, emphasizing their role in system controllability. Similarly, (Ibarra-Sánchez and Segovia-Hernández, 2010) have addressed operational condition optimization from a control perspective, striving to maximize not only energy efficiency but also the controllability of the system. These studies represent a diverse yet interwoven body of research, centrally focused on the controllability of bioethanol extractive distillation. Expanding the scope, the work of (Wolf Maciel and Brito, 1995) has provided insights into the control dynamics of multi-component bioethanol systems, enriching our understanding of controllability challenges in more complex scenarios. Additionally, the investigations of (Kong et al., 2022) have offered a comprehensive exploration of advanced control algorithms, contributing to the arsenal of tools available for improving the controllability of bioethanol purification processes. Collectively, this wealth of research, integrating open-loop and closed-loop approaches, not only refines our understanding of bioethanol extractive distillation controllability but also lays the foundation for the development of sophisticated control strategies. It sets the stage for further exploration, emphasizing the critical interplay between system design and controllability in advancing the efficiency and reliability of bioethanol production processes.

The goal of this study was to develop a purification process for dehydrating bioethanol through extractive distillation. This process was specifically designed for an ethanol-water mixture obtained from the fermentation of sugars derived from the transformation of sotol bagasse, as previously published by these authors. (González-Chavez et al., 2022). The aforementioned study focused on experimenting with the essential phases of the process and designing reactors for pretreatment, enzymatic hydrolysis, and fermentation; however, the distillation sequence was not taken into account. In that sense, to the best of our knowledge, there is no existing literature on the investigation of a distillation process and the study of its dynamic properties for processing an azeotropic mixture of ethanol and water derived from the fermentation of sugars obtained from sotol bagasse. Finally, the process controllability was evaluated in order to find an efficient and controlled system for the specific purification process of bioethanol from SB. In this research, control properties were analyzed using the singular value decomposition technique and closed loop analysis. Herein, we provide insights about the dynamic performance of this complex system.

## 2. Methodology

## 2.1. Design strategy

We present an energy-efficient design of extractive distillation. In order to overcome the complexity that represents the simultaneous solution of the tray arrangement and energy consumption within a formal optimization algorithm, we decoupled the design problem in two stages: (i) tray configuration and (ii) optimal energy consumption. The distillation sequence considers a depletion column, an extractive column (with glycerin as a solvent agent) and a solvent recovery column (Fig. 1). The columns were calculated using the Non-Random Two Liquids model (NRTL). The NRTL method is indeed specifically designed to predict activity coefficients in non-ideal liquid mixtures. The ethanol-water mixture is an example of a non-ideal solution due to differences in molecular interactions, particularly hydrogen bonding. When dealing with binary systems, such as ethanol-water, the NRTL method is an effective choice for modeling the non-ideal behavior of the mixture. This method can describe non-idealities in the distribution of molecules in liquid phases and can provide more accurate predictions of activity coefficients and thermodynamic properties compared to simpler models based on Raoult's law (Avilés Martínez et al., 2012; Gebreyohannes et al., 2014; Pacheco-Basulto et al., 2012).

In the first stage of our approach, preliminary designs for the extractive system were developed using different parameters considered in conventional distillation columns such as temperature and pressure. After the tray arrangement for the extractive sequence was obtained, an optimization procedure minimized the heat duty supplied to the reboilers of the complex scheme. In this case, the required purity constraints of the three streams were considered. Subsequently, the degrees of freedom that remain after design specifications and tray arrangement were used to obtain the operating conditions that provide minimum energy consumption. In the complex sequence, two degrees of freedom remained: the side solvent flow and the solvent stream stage.



Fig. 1. Extractive distillation sequence.

The optimization strategy can be summarized as follows: (a) a base design scheme is obtained; (b) values for the solvent stream are assumed; (c) a rigorous model for the simulation of the complex scheme with the proposed tray arrangement is solved. For this purpose, Aspen Plus<sup>TM</sup> V10 was used in the present study. In the event product compositions are obtained, the design is maintained; otherwise, appropriate adjustments are performed; (d) process is repeated until a local minimum in energy consumption for the assumed values are detected; (e) the value for the stream stage is modified and step (c) is repeated until the energy consumption reaches its minimum value. This result provides the optimal value that should be included in the design of the complex scheme. This optimization methodology is the one proposed in (Gutiérrez-Guerra et al., 2009), (Sánchez-Ramírez et al., 2020), among others.

## 2.2. Dynamic analysis

The execution of closed-loop and open-loop studies in chemical process control extends beyond the mere presence of dynamic simulation tools like Aspen Dynamics, as it provides a comprehensive and indepth understanding of a system's dynamic behavior. These investigations are critical for various additional reasons. Firstly, closedloop studies enable the validation of dynamic models used in simulations, such as those implemented in Aspen Dynamics. By comparing real-time system responses with model predictions, the precision of the dynamic model can be verified and adjusted. Secondly, closed-loop studies are instrumental in optimizing controller design by revealing how controllers respond to changes in process conditions, thereby enhancing overall stability and performance. Thirdly, the analysis of system stability is crucial in practical applications, and closed-loop studies allow for the evaluation of system stability under dynamic conditions, assessing controller robustness against disturbances and variations in operating conditions. Moreover, closed-loop studies assist in detecting potential operational issues and improving the system's ability to recover from undesirable conditions. Additionally, they provide a simulated environment for operator training, enhancing operators' skills and decision-making in real-life situations. Lastly, the combination of closed-loop and open-loop studies facilitates ongoing research and the development of advanced control strategies, ensuring continuous improvement in operational efficiency, regulatory compliance, and adaptation to changes in the environmental conditions. In summary, the implementation of closed-loop and open-loop studies is essential to ensure optimal performance and reliability in chemical process control, encompassing model validation, controller optimization, stability analysis, issue detection, operator training, and continuous research and development.

#### 2.2.1. Singular value decomposition (SVD)

Open loop dynamic responses to set point changes around the assumed operating point were achieved. The responses were obtained through the use of Aspen Plus<sup>TM</sup>. Transfer function matrices (G) (Eq. 1) were then collected for each case, and they were subjected to singular value decomposition (SVD):

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

where  $\Sigma = \delta \iota \alpha \gamma$  ( $\sigma_1$ ,  $\Box$ ,  $\Box$ ,  $\sigma_\nu$ ),  $\sigma_i$  = singular value of  $G = \lambda_i^{1/2}$  (GG<sup>H</sup>); V = (v<sub>1</sub>, v<sub>2</sub>, ...) matrix of left singular vectors, and W = (w<sub>1</sub>, w<sub>2</sub>,...) matrix of right singular vectors. The two parameters of interest included the minimum singular value,  $\sigma_*$ , and the ratio maximum to minimum singular values or condition number (Eq. 2):

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

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 when uncertainties in process parameters and modeling errors occur. These parameters provide a qualitative assessment of the theoretical control properties of the alternate designs. The systems with the highest minimum singular values and lowest condition numbers are expected to show the best dynamic performance under feedback control. A full SVD analysis should cover a sufficiently complete range of frequencies. For this initial analysis of the extractive configurations, we estimated the SVD properties for each separation system at zero frequency. Such analysis should provide preliminary indication on the control properties of each system. A similar analysis has reported by Alcocer-García et al. (2020).

#### 2.2.2. Closed-loop simulations

To supplement the SVD analysis, rigorous dynamic simulations under closed loop operation were carried out. For the closed-loop analysis, several conditions are defined. They include the control loops for each system, the type of process controller to be used, and the values of the controller parameters. Several techniques including the relative gain array method can be used to fix the loops present in a control system. However, in the case of distillation columns, the loops are well established. From a practical point of view, they are successfully at least in conventional columns. A well-known structure is based on energy balance considerations, which yields the so-called LV control structure where 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 compositions of the distillate and bottom outputs (Haggblom et al., 1992; Yang et al., 2019). The control loops for the integrated systems were obtained from different parameters generated in conventional distillation columns. The objective of the control stage was to preserve the specified purity in the output streams of the studied design. Two control composition loops are identified after operation of conventional columns. For the control of the top product stream, the reflux flowrate was used. In addition, for the control of the bottom product stream, the reboiler heat duty was chosen. It is worth mentioning that the selected control loops have been used with satisfactory results in previous studies conducted on thermally coupled systems (Jiménez et al., 2001; Segovia-Hernández et al., 2002; Segovia-Hernández et al., 2004). The selection of the type of controller considered that the Proportional-Integral (PI) mode is widely used in distillation systems in the industry. The choice may also provide a basis for the comparison of more elaborated control laws. In the selection of the of the PI controllers parameters, care was taken to provide a common method for each of the compared sequences. A tuning procedure that involved the minimization of the integral of the absolute value (IAE) for each loop of each scheme was used (Stephanopoulos, 1984). Therefore, for each loop, an initial value of the proportional gain was established and a search over the values of the integral reset time was conducted until the local optimum value of the IAE was obtained  $\left(\int_0^T |e(t)| dt\right)$ . The term T is the upper limit which is selected arbitrarily. The process was repeated for other proportional gain values. The controller parameters set that provided a global minimum value of the IAE was selected. Although the tuning procedure was properly elaborated, the control analysis was conducted based on a common tuning method for the controller parameters. In the context of chemical process control, the integral of absolute error (IAE) stands as a quantitative metric employed to assess dynamic characteristics within a control system. The IAE is a crucial performance measure that calculates the accumulated sum of absolute values of the error signal over a defined time interval. The error signal reflects the disparity between the desired setpoint and the actual process variable, capturing the extent of deviation from the intended system behavior. The dynamic characteristics elucidated by the IAE provide insights into the system's efficacy in accurately tracking and regulating the specified setpoint. A lower IAE value denotes minimal deviation and tighter control, indicating superior dynamic performance. Conversely, a higher IAE value suggests a more significant cumulative error, signifying

challenges in maintaining the desired setpoint and potential inefficiencies in the applied control strategy. Fundamentally, the IAE offers a quantitative lens through which to analyze the dynamic behavior of a chemical process control system by measuring the cumulative impact of deviations over time. This metric serves as a valuable tool for engineers and researchers, facilitating the evaluation of control algorithm effectiveness, the tuning of controller parameters, and the optimization of the overall dynamic performance of the system.

The primary aim of the simulations was to assess the dynamic behavior of the systems operating in feedback control mode. As part of the case study, servo control was specifically chosen to conduct a closedloop analysis. In this analysis, deliberate step changes were introduced into the setpoint for each product composition, all of which were subject to Single Input Single Output (SISO) feedback control. This experimental setup allowed for a comprehensive examination of the dynamic response of the systems to changes in setpoint conditions, particularly focusing on the servo control strategy. The induced step changes in the setpoint for each product composition provided a nuanced perspective on the systems' adaptive capabilities and response dynamics under the influence of SISO feedback control. The integration of these deliberate setpoint variations further enriched the study by offering insights into how the systems navigate and adapt to altered operating conditions. Through this approach, the evaluation of dynamic behavior extended beyond a generic assessment, providing a more specific and detailed understanding of the systems' performance under the chosen feedback control strategy.

#### 2.3. Case study

Previously, our research group published the experimental transformation of SB into bioethanol (González-Chavez et al., 2022). The train used in that work consisted of a pretreatment, enzymatic hydrolysis, and fermentation step. However, the distillation stage of the fermented broth was not evaluated. Herein, we propose the use of a three-column system to obtain ethanol with a purity > 99.9% (See Fig. 1).

The purification train described in this research has been based on that previously published by (Errico et al., 2015). The purification setup comprises three columns designed for obtaining high ethanol purity. The first column, functioning as a stripping unit, removes the majority of water from the low-concentration ethanol mixture resulting from fermentative processes. The distillate from the stripping column is directed to an extractive column, where a solvent aids in breaking the azeotrope and recovering ethanol in a highly pure form. Ethylene glycol (EG) was used as the solvent agent at ratios of 1.5:1, 2:1, and 2.5:1. EG was selected because it helps reduce the energy consumption required to separate the ethanol-water mixture. It is crucial to emphasize that the specified values for the solvent-to-feed flow rate ratio are standard in industrial practice and extensively documented in the literature (Gutiérrez-Guerra et al., 2009). This has been reported by various authors (Gil et al., 2014; Ravagnani et al., 2010). However, EG has the disadvantage of having higher toxicity. Later, the dynamic analysis previously described in Section 2 was used to determine the controllability of the process. Finally, the solvent, now mixed with water, exits the extractive column, entering a third column for precise separation. The purified solvent is then recycled for reuse in the extraction column. This paper presents a novel approach to extractive distillation, emphasizing that while the configuration employed in this study mirrors previous works, the novelty lies in the composition of the initial mixture. This composition, derived from a previously unexplored raw material, adds a unique dimension to the study. The chosen case study, unanalyzed in existing literature, further contributes to the originality and significance of this research.

This design was previously optimized to reduce energy consumption. The characteristic of each column is presented in Table 1 and Fig. 2 presents the controller diagram.

Table 1
Feed characterization

Mole flow	82,866.100
Pressure (kPa)	8.900
Vapor fraction	0
Enthalpy (kcal)	82,866.145
Composition (mass fraction)	
Ethanol	0.054
Glucose	0.007
Xylose	0.005
Water	0.934

#### 3. Results

The distillation sequences evaluated in the present research considered three different feed stream:solvent ratios. The results are presented herein. Table 2 displays the parameters and energy consumption of the design corresponding to the extractive distillation sequence with a solvent ratio of 2:1 in the feed stream. This sequence resulted in optimal control characteristics, signifying that systems with the highest minimum singular values and the lowest condition numbers are anticipated to exhibit superior dynamic performance under feedback control. Furthermore, it is expected that these systems will demonstrate the least Integral of Absolute Error (IAE) values. In both open-loop and closedloop scenarios for examining the dynamic performance of the process, disturbances introduced were set at 1%, following the recommendation by Alcocer-García et al. (2020).

Furthermore, it is anticipated that these systems will exhibit the lowest Integral of Absolute Error (IAE) values. In the investigation of dynamic performance in both open-loop and closed-loop scenarios, the obtained results offer a comprehensive analysis that sheds light on various aspects of the distillation process.

The design parameters for the distillation system columns are presented in Table 2. Notably, the MR column stands out as the most energy-intensive, with energy consumption in the reboiler being 8 and 20 times higher than that of the DE and DR columns, respectively. This is associated with the need to remove large quantities of water from the fermentation liquors.

This results in a significantly high reflux ratio for the MR column, attributed to the substantial water removal required in the initial column. The recirculation process enhances purity in the early stages. In contrast, the DE and DR columns experience a drastic reduction in reflux ratio, likely due to the higher purity of mixtures in these columns. Additionally, the column diameters follow the order MR>DE>DE, reflecting the larger diameter of the MR column, which is associated with the substantial water removal needed in this particular column.

In Fig. 3, which portrays the composition of the ethanol-water mixture at different stages of the columns, notable observations emerge. Fig. 3(a) highlights the MR column's exceptional water removal capabilities, as evidenced by a bottom composition registering 98.77% water. Meanwhile, Fig. 3(b) unveils an impressive ethanol concentration of 99.91% at the top of the extractive column, resulting in a peak ethanol recovery of 4472.65 kg ethanol per hour. Moving to the DR column (Fig. 3(c)), the solvent agent achieves a remarkable recovery, culminating in a final purity of 99.97%. This high-purity compound is subsequently recirculated to the DE extractive column, illustrating the efficiency of the distillation process.

Examining the composition of ethanol as influenced by the dynamic response in the recovery and extractive distillation columns (Fig. 4), the oscillatory behavior inherent in controllability processes is evident across all three sequences. As reported in previous studies (Errico et al., 2015), oscillations were notably suppressed at 0.6 h when the glycerol: water ratio was 2.5:1. Additionally, oscillations ceased at 1.5 h for solvent: water ratios of 1.5:1 and 2:1, showcasing the dynamic response's sensitivity to these specific operational conditions.

Table 3 reinforces these findings, indicating that the IAE value



Fig. 2. Controller diagram.

### Table 2

Design parameters and energy consumption of the extractive distillation sequence.

	MR	DE	DR
Total stages	15	20	8
F stage number	10	12	4
Reflux ratio	14.9038	1.1	1
Solvent f stage		3	
Column Press (bar)	1.01325	1.01325	1.01325
$\Delta P$ (bar)	0.16	0.23	0.15
Diameter (m)	3.2429	0.768116	1.04013
D flow rate (kg/hr)	5226.08	4472.65	755.452
Water 1 f rate (kg/hr)	76,687.9		
Water 2 f rate (kg/hr)			753.539
Solvent flow rate		10,447.90	
Purity bioethanol (%wt)		0.9991	
Purity water 1 (%wt)	0.9877		
Purity water 2 (%wt)		0.9975	
Purity solvent recycle (%wt)			0.9997
Condenser (kW)	-24559.8	-2225.94	-949.501
Reboiler (kW)	26,800.5	3249.49	1313.17
Total condenser	-27735.241		
Total reboiler	31,363.16		

corresponds to the 2.5:1 solvent ratio, validating the trends observed in Fig. 4. Beyond mere numerical values, this data underscores the nuanced interplay between process parameters and dynamic behavior, offering insights into the controllability landscape of the distillation system.

An intriguing aspect arises when considering the impact of gain parameters (Kc) on controllability. Remarkably, the highest Kc value is observed at the 2.5:1 ratio, aligning with the research by (Errico et al., 2015) and (Ramírez-Márquez et al., 2013a). These studies noted that increased Kc values correlated with IAE minimization, indicating a favorable improvement in controllability. This correlation emphasizes the importance of not only understanding dynamic responses but also leveraging this understanding to optimize control strategies for enhanced system performance.

It is imperative to underscore that the oscillatory patterns discerned in the control process, as illustrated in this figure, are consistent with a distinct and noteworthy characteristic: the well-documented oscillatory behavior observed in earlier studies, notably by (Errico et al., 2015). The complexity of these oscillations is intricately tied to the unique presence of an azeotrope within the distillation process. In essence, the azeotrope—a mixture of components that evaporates without a change in composition—introduces a level of intricacy to the system's dynamic response. Furthermore, the solvent's role in this scenario becomes paramount, as it exerts a discernible influence on the azeotrope, contributing to its breakdown and subsequent impact on the dynamic behavior of the entire distillation process. This nuanced interplay between the azeotrope and the solvent amplifies the complexity of the system's controllability, rendering the observed oscillations a consequential manifestation of the intricate dynamics inherent in the distillation of such complex mixtures. This insight not only advances our understanding of the oscillatory behavior in distillation processes but also underscores the importance of considering azeotropes and solvent effects in the design and control of these systems.

It is crucial to highlight the delicate equilibrium between controllability and energy consumption. In this context, the optimal design is



Fig. 3. Composition profile of the: a) MR; b) DE; and c) DR columns.



Fig. 4. Dynamic response for extractive distillation column DE.

#### Table 3

Optimum closed loops LV for the three solvent ratios evaluated in the extractive column.

Column	Solvent ratio 2:1			
	Kc	$\tau_i$ (h)	IAE	
MR	249	20	0.00350493	
DE	155	20	0.00336184	
DR (top)	250	20	0.00334655	
DR (bottoms)	248	20	0.00338451	
Column	Solvent ratio 2.5:1			
	Kc	$\tau_i$ (h)	IAE	
MR	250	20	0.00350420	
DE	250	20	0.00335778	
DR (top)	250	20	0.00334293	
DR (bottoms)	248	20	0.00338954	
Column	Solvent ratio 1.5:1			
	Kc	$\tau_i$ (h)	IAE	
MR	249	20	0.00350493	
DE	50	20	0.00343854	
DR (top)	250	20	0.00334832	
DR (bottoms)	250	20	0.00337027	

characterized by achieving both low energy consumption and high controllability. Our findings reveal that the sequence utilizing a solvent: feed ratio of 1.5:1 displayed lower energy consumption compared to ratios of 2:1 and 2.5:1. However, the 1.5:1 ratio exhibited lower controllability, while the other two ratios demonstrated higher controllabilities. This observation underscores the inherent trade-off in process design and control strategy selection, prompting careful consideration of these factors in practical applications.

Furthermore, the analysis discerns that the solvent: feed ratio of 2.5:1 not only provides the lowest IAE value but also achieves a high Kc. Among the three ratios studied, the 2:1 ratio displayed the most stable profile, indicative of its robust dynamic response. Therefore, to strike an optimal balance between energy efficiency and controllability, the sequence with a solvent:feed ratio of 2:1 emerges as a promising candidate for further exploration in subsequent studies. The insights gained from this comprehensive analysis contribute not only to a deeper understanding of the dynamic behavior of the distillation process but also serve as a valuable guide for refining control strategies and optimizing energy consumption in industrial applications.

#### 4. Conclusions

In the context of this investigation, a distillation system tailored for ethanol purification was thoroughly examined. Specifically, focus was directed toward an ethanol-water mixture derived from the conventional biochemical processing of SB. The application of an extractive distillation scheme, utilizing ethylene glycol as the solvent agent, resulted in the achievement of an impressive ethanol purity of 99.91%. Controllability was a pivotal aspect of the study, and three distinct solvent-to-feed ratios (1.5:1, 2:1, and 2.5:1) were systematically evaluated. The data highlighted that optimal controllability was attained at a solvent-to-feed ratio of 2.5:1, while the lowest energy consumption was observed at a ratio of 1.5:1. Notably, the sequence employing a solventto-feed ratio of 2:1 emerged as the most balanced, offering an equilibrium between controllability and energy savings. Furthermore, this equilibrium was maintained under these conditions, reinforcing the robustness of the selected design. Consequently, the conclusion drawn is that the distillation sequence utilizing a solvent-to-feed ratio of 2:1 represents an ideal compromise between controllability and energy efficiency. This positioning identifies it as a preferred choice for further modifications or potential implementation in industrial settings, given its ability to strike a harmonious balance between energy consumption and control properties.

## **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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#### References

- Alcocer-García, H., Segovia-Hernández, J.G., Prado-Rubio, O.A., Sánchez-Ramírez, E., Quiroz-Ramírez, J.J., 2020. Multi-objective optimization of intensified processes for the purification of levulinic acid involving economic and environmental objectives. Part II: a comparative study of dynamic properties. Chem. Eng. Process. Process. Intensif. 147 https://doi.org/10.1016/j.cep.2019.107745.
- Arifin, S., Chien, I.L., 2008. Design and control of an isopropyl alcohol dehydration process via extractive distillation using dimethyl sulfoxide as an entrainer. Ind. Eng. Chem. Res. 47, 790–803. https://doi.org/10.1021/ie070996n.
- Avilés Martínez, A., Saucedo-Luna, J., Segovia-Hernandez, J.G., Hernandez, S., Gomez-Castro, F.I., Castro-Montoya, A.J., 2012. Dehydration of bioethanol by hybrid process liquid-liquid extraction/extractive distillation. Ind. Eng. Chem. Res. 5847–5855. https://doi.org/10.1021/ie200932g.
- Errico, M., Ramírez-Márquez, C., Torres, E., Rong, B., Segovia-Hernández, J.G., 2015. Design and control of an alternative distillation sequence for bioethanol purification. J. Chem. Technol. Biotechnol. 90, 2180–2185. https://doi.org/10.1002/jctb.4529.
- Flores-Gallegos, A.C., Cruz-Requena, M., Castillo-Reyes, F., Rutiaga-Quiñones, M., Sepulveda-Torre, L., Paredes-Ortíz, A., Soto, O., Rodriguez-Herrera, R., 2019. Sotol, an alcoholic beverage with rising importance in the worldwide commerce. Alcoholic Beverages. Elsevier Inc. https://doi.org/10.1016/B978-0-12-815269-0.00005-2. Gebrevohannes, S., Neely, B.J., Gasem, K.A.M., 2014. One-parameter modified
- nonrandom two-liquid (NRTL) activity coefficient model.
- Gil, I.D., García, L.C., Rodríguez, G., 2014. Simulation of ethanol extractive distillation with mixed glycols as separating agent. Braz. J. Chem. Eng. 31, 259–270.
- Gil, I.D., Gómez, J.M., Rodríguez, G., 2012. Control of an extractive distillation process to dehydrate ethanol using glycerol as entrainer. Comput. Chem. Eng. 39, 129–142. https://doi.org/10.1016/j.compchemeng.2012.01.006.
- González-Chavez, J. de J., Arenas-Grimaldo, C., Amaya-Delgado, L., Vázquez-Núñez, E., Suarez-Vázquez, S., Cruz-López, A., Segovia-Hernández, J.G., Pérez-Vega, S., Salmerón, I., Molina-Guerrero, C.E., 2022. Sotol bagasse (Dasylirion sp.) as a novel feedstock to produce bioethanol 2G: bioprocess design and biomass characterization. Ind. Crops Prod. 178, 0–1. https://doi.org/10.1016/j.indcrop.2022.114571.
- Gutiérrez-Guerra, R., Segovia-Hernández, J.G., Hernández, S., 2009. Reducing energy consumption and CO<sub>2</sub> emissions in extractive distillation. Chem. Eng. Res. Des. 87, 145–152. https://doi.org/10.1016/j.cherd.2008.07.004.

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Haggblom, K.E., Waller, K.V., Akademi, A., 1992. In: Luyben, W.L. (Ed.), Control Structures, Consistency, and Transformations. Practical Distillation Control, New York, NY, pp. 192–193.

- Hernández-Vargas, E.A., Sánchez-Ramírez, E., de la Fuente, A.R., Calderón-Alvarado, M. P., Segovia-Hernández, J.G., 2023. Advancing neural network architectures for time series forecasting: a sustainable approach to intensified biobutanol production. Chem. Eng. Process. Process. Intensif. 194, 109603 https://doi.org/10.1016/j. cep.2023.109603.
- Ibarra-Sánchez, J. de J., Segovia-Hernández, J.G., 2010. Reducing energy consumption and CO<sub>2</sub> emissions in extractive distillation: part II. Dynamic behavior. Chem. Eng. Res. Des. 88, 135–145. https://doi.org/10.1016/j.cherd.2009.08.006.
- Jiménez, A., Hernández, S., Montoy, F.A., Zavala-García, M., 2001. Analysis of control properties of conventional and nonconventional distillation sequences. Ind. Eng. Chem. Res. 40, 3757–3761. https://doi.org/10.1021/ie000047t.
- Kong, Z.Y., Sánchez-Ramírez, E., Yang, A., Shen, W., Segovia-Hernández, J.G., Sunarso, J., 2022. Process intensification from conventional to advanced distillations: past, present, and future. Chem. Eng. Res. Des. 188, 378–392. https:// doi.org/10.1016/j.cherd.2022.09.056.
- López-Ortega, M.G., Guadalajara, Y., Junqueira, T.L., Sampaio, I.L.M., Bonomi, A., Sánchez, A., 2021. Sustainability analysis of bioethanol production in Mexico by a retrofitted sugarcane industry based on the Brazilian expertise. Energy 232, 121056. https://doi.org/10.1016/j.energy.2021.121056.
- Molinuevo-Salces, B., Riaño, B., Hijosa-Valsero, M., González-García, I., Paniagua-García, A.I., Hernández, D., Garita-Cambronero, J., Díez-Antolínez, R., García-González, M.C., 2020. Valorization of apple pomaces for biofuel production: a biorefinery approach. Biomass Bioenergy 142. https://doi.org/10.1016/j. biombioe.2020.105785.
- Oseguera-Villaseñor, I., Martínez-Rodríguez, G., Barroso-Muñoz, F.O., Segovia-Hernández, J.G., Hernández, S., 2018. Multiplicities in dividing wall distillation columns in the purification of bioethanol: energy considerations. Clean. Technol. Environ. Policy 20, 1631–1637. https://doi.org/10.1007/s10098-017-1415-0.
- Pacheco-Basulto, J. ángel, Hernández-McConville, D., Barroso-Muñoz, F.O., Hernández, S., Segovia-Hernández, J.G., Castro-Montoya, A.J., Bonilla-Petriciolet, A., 2012. Purification of bioethanol using extractive batch distillation: simulation and experimental studies. Chem. Eng. Process. Process. Intensif. 61, 30–35. https://doi.org/10.1016/j.cep.2012.06.015.
- Pino, M.S., Rodríguez-Jasso, R.M., Michelin, M., Flores-Gallegos, A.C., Morales-Rodriguez, R., Teixeira, J.A., Ruiz, H.A., 2018. Bioreactor design for enzymatic hydrolysis of biomass under the biorefinery concept. Chem. Eng. J. 347, 119–136. https://doi.org/10.1016/j.cej.2018.04.057.
- Piñón-Muñiz, M.I., Ramos-Sánchez, V.H., Gutiérrez-Méndez, N., Pérez-Vega, S.B., Sacramento-Rivero, J.C., Vargas-Consuelos, C.I., Martinez, F.M., Graeve, O.A., Orozco-Mena, R.E., Quintero-Ramos, A., Sánchez-Madrigal, M.A., Salmerón, I., 2023. Potential use of Sotol bagasse (Dasylirion spp.) as a new biomass source for

liquid biofuels production: comprehensive characterization and ABE fermentation. Renew. Energy 212, 632–643. https://doi.org/10.1016/j.renene.2023.05.055.

- Ramírez-Márquez, C., Segovia-Hernández, J.G., Hernández, S., Errico, M., Rong, B.G., 2013a. Dynamic behavior of alternative separation processes for ethanol dehydration by extractive distillation. Ind. Eng. Chem. Res. 52, 17554–17561. https://doi.org/10.1021/ie402834p.
- Ramírez-Márquez, C., Segovia-Hernández, J.G., Hernández, S., Errico, M., Rong, B.G., 2013b. Dynamic behavior of alternative separation processes for ethanol dehydration by extractive distillation. Ind. Eng. Chem. Res. 52, 17554–17561. https://doi.org/10.1021/ie402834p.
- Ravagnani, M.A.S.S., Reis, M.H.M., Filho, R.M., Wolf-Maciel, M.R., 2010. Anhydrous ethanol production by extractive distillation: a solvent case study. Process Saf. Environ. Prot. 88, 67–73. https://doi.org/10.1016/j.psep.2009.11.005.
- Sanchez, A., Sevilla-Güitrón, V., Magaña, G., Gutierrez, L., 2013. Parametric analysis of total costs and energy efficiency of 2G enzymatic ethanol production. Fuel 113, 165–179. https://doi.org/10.1016/j.fuel.2013.05.034.
- Sánchez-Ramírez, E., Ramírez-Márquez, C., Quiroz-Ramírez, J.J., Angelina-Martínez, A. Y., Vicente Cortazar, V., Segovia-Hernández, J.G., 2020. Design of dividing wall columns involving sustainable indexes for a class of quaternary mixtures. Chem. Eng. Process. Process. Intensif. 148 https://doi.org/10.1016/j.cep.2020.107833.
- Segovia-Hernández, J.G., Hernández, S., Jiménez, A., 2002. Control behaviour of thermally coupled distillation sequences. Trans. IChemE 80, 783–789.
- Segovia-Hernández, J.G., Hernández, S., Rico-Ramírez, V., Jiménez, A., 2004. A comparison of the feedback control behavior between thermally coupled and conventional distillation schemes. Comput. Chem. Eng. 811–819. https://doi.org/ 10.1016/j.compchemeng.2004.02.019.
- Stephanopoulos, G., 1984. Chemical Process Control: An Introduction to Theory and Practice. Englewood Cliffs, NY.
- Torres-Ortega, C.E., Ramírez-Márquez, C., Sánchez-Ramírez, E., Quiroz-Ramírez, J.J., Segovia-Hernandez, J.G., Rong, B.G., 2018. Effects of intensification on process features and control properties of lignocellulosic bioethanol separation and dehydration systems. Chem. Eng. Process. - Process. Intensif. 128, 188–198. https:// doi.org/10.1016/j.cep.2018.04.031.
- Vázquez-Ojeda, M., Segovia-Hernández, J.G., Hernández, S., Hernández-Aguirre, A., Kiss, A.A., 2013. Design and optimization of an ethanol dehydration process using stochastic methods. Sep Purif. Technol. 105, 90–97. https://doi.org/10.1016/j. seppur.2012.12.002.
- Wolf Maciel, M.R., Brito, R.P., 1995. Evaluation of the dynamic behaivor of an extractive distillation column for dehydration of aqueuos ethanol mixtures. Comiputers Them. Engng.
- Yang, A., Shen, W., Wei, S., Dong, L., Li, J., Gerbaud, V., 2019. Design and control of pressure-swing distillation for separating ternary systems with three binary minimum azeotropes. AIChE J. 65, 1281–1293. https://doi.org/10.1002/aic.16526.