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Towards sustainability assessment through a flexibility index as the condition number

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ABSTRACT

Green chemistry and green engineering, projected in the concept of sustainability, provide inherently safe processes. These safety aspects can be determined indirectly through process controllability and flexibility. Although several indexes have been proposed to measure these parameters, the flexibility index and controllability measured by the condition number are notable. However, these two indicators are usually measured individually, and information inherent to synergies is lost due to the link between profitability and process safety. In the present work we aim to evaluate a quantitative parameter (from the condition number), to simultaneously measure flexibility and controllability of processes to generate a simple measure of sustainability to processes. To demonstrate the relationship of controllability and flexibility, through the condition number, three sequences of solar grade silicon production were explored. In addition, the advantages of using the proposed indicator were demonstrated through a pair of binary and multicomponent reactive distillation sequences. The results clearly showed that the condition number measuring controllability at low frequencies can be related to flexibility. And that this is potentially an indicator that can simultaneously measure controllability and flexibility, and with its use in process design, sustainable processes will be ensured.

1. Introduction

The need for efficient and sustainable processes in the chemical industry is becoming increasingly evident. With processes that meet these characteristics, the main challenges of today, such as energy consumption, resource depletion and environmental impact, will be confronted. The inclusion of distinctive features such as cost-effectiveness, safety, operability, and environmental footprint at an early design stage are essential to address the needs. Wang et al., [1] state that the development of efficient and sustainable processes is paramount for the chemical industry to remain competitive. Furthermore, one of the primary objectives is the advancement of inherently safe processes with minimal environmental impact. Cheng and Zhao [2] propose that to achieve efficient and sustainable processes, systems that are safe and reliable are required. But how can safe and reliable processes be obtained? The answer lies in achieving processes with good controllability and flexibility. These two measures are an indirect indicator of a safe and reliable process. Therefore, having an indicator that can evaluate the controllability of a system, as well as its flexibility, and that simultaneously and indirectly quantifies objectives such as inherent safety, economy, or environmental impact, will be very useful for the design and optimization of chemical processes. Specifically, having such an indicator improve the inherent safety of any process would favor a more effective risk management. Actions can be taken to avoid accidents rather than mitigate their consequences [3].

The indicator could contribute to the industrial processes that cover the characteristics of green chemistry and green engineering. According to the concepts exposed by Jiménez-González and Constable [4], green chemistry and green engineering should be considered an integral part of broader context of sustainability. In fact, in green chemistry, aspects such as safety, process control, health and environmental impact are often highlighted. The eleventh principle of green chemistry expresses the aspiration for real-time process analysis and traceability. The objective of this principle is to prevent safety and waste issues by identifying process disturbances in real time. If any of these issues were to occur, time to adjust the process parameters or dampen the disturbance should be available [4]. Therefore, if perturbations occur, there

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Nomenclature

τ	Time's constants
J_A^k	Sets of the $n+1$ restrictions involved in the kth active set, k=1, …, n_{AS}
λ_j^k	Constants
σ_*	Minimum singular value
σ^*	Maximum singular value
D	Diameter [m]
Eco99	Eco-indicator 99 [P/y]
IR	Individual Risk [1/y]
k	Process gain
MINLP	Mixed integer nonlinear programming
Q	Reboiler duty [cal/sec]
RI	Resilient Index
ROI	Return on Investment [%]
ω	Frequency [rad/s]
γ *	Condition number
θ_d	Dead time

must be sufficient time for the process to effectively cope and impact positively not only into the safety, but also in the final product quality. The evaluation of system controllability, together with the evaluation of process flexibility, may contribute to be tools to drive sustainability from a triple bottom line perspective with influence on the aspects of sustainability. Works from Palma-Barrera et al. [5] and Contreras-Zarazúa et al., [6] demonstrate that with the evaluation of the condition number as a control indicator, it is possible to have sustainable and inherently safe processes.

As it has been mentioned, flexibility and control are parameters with a great impact on process design and optimization. Although they could be thought of as similar concepts, control and flexibility correspond to different technical notions. The optimal operation of a process in the presence of disturbances could be maintained using optimization-based control [7], which makes control one of the most important considerations, along with flexibility, to maintain operation plant optimum, which is related to the quality and stability of the dynamic response of a process. A quantitative measure frequently used in the measurement of control is the condition number [8]. The condition number represents the stability and quality of the process in a dynamic state [9]. Flexibility indicates that the operation of the process is feasible under different steady-state operating conditions [10]. The condition number provides valuable information in theoretical control for a given frequency in a transfer function matrix in a multivariate system. So, the condition number ends up being the ratio between the maximum and minimum singular value and represents the sensitivity of the system to the uncertainty present at the input. Therefore, it is hypothetically possible that the condition number can be used as an indirect measure of the flexibility of a process.

From the perspective of process optimization, the inclusion of a single integrated indicator is of great value to obtain sustainable processes from the design phase. There are many works that optimize processes [11–13], and that multi-objectively evaluate the objective functions of: economy, inherent safety, environmental impact, and/or controllability. This represents a great computational effort, which is reflected in a long optimization time [14]. Therefore, with a single indicator such as the condition number, capable of evaluating all the above, the optimization time of a process could be reduced. Cabrera-Ruiz et al., [15] had previously presented the integration of the condition number as a controllability criterion in a global stochastic optimization algorithm that can solve single-objective or multi-objective optimization problems. In their work, the possibility of assessing the controllability of distillation sequences using only steady-state information is visualized. The results showed that simplified process dynamics can address the optimal design and control of intensified distillation sequences. Pioneering the inclusion of a controllability indicator in process optimization. Later Cabrera-Ruiz et al., [16] proposed an extended version of the condition number evaluation from a formal dynamic state evaluation of the intensified processes.

Di Pretoro et al., [17] have defined a procedure for the evaluation of flexibility and have compared the different flexibility indexes found in the literature to perform a simple evaluation of flexibility in distillation schemes. They have also made a first attempt to associate flexibility with economic aspects of distillation sequences which result in a direct relationship between lower investment cost and a higher process flexibility. Di Pretoro et al., [18] differentiate the performance of different integrated and conventional unit designs under uncertain conditions. This was done for different ranges of flexibility which has allowed them to select the most suitable configuration for the required performance. Their expectations on investment costs and emissions are more thoroughly calculated when flexibility is considered. And finally, Ochoa et al., [19] propose new MINLP reformulations for the extended flexibility analysis. They have distinguished an efficient way of calculating a new flexibility index for the process parameters.

Di Pretoro et al., [20] made an analysis with a distillation train where the flexibility could be of critical to assess the operating conditions range of better performance for different system configurations. In this article the comparison between a weak and strong flexibility constraints and resulted in a dedicate additional costs vs. flexibility trend useful to improve the decision making.

Having furthermore, works where the controllability is introduced as a degree of freedom along with minimizing operating costs, as Halvorsen and Skogestad [21] presents on feedback control of divided wall columns, where they identified 5 degrees of freedom, 3 of them controlled variables, and the two additional for minimizing the cost. Wolf and Skogestad [22] describe a Petlyuk column control, they found that in a 3 components case, there are no control problems. Nowadays, there are also studies that proposed new methods to process industry to enhance its operational efficiency. Shin et al., [23] used an artificial neural network model to improve the model predictive control. Showing in the results a better performance in a biorefinery distillation train.

However, none of these works or any others presented in the literature show the condition number as a quantitative measure of the flexibility of a process.

The novelty of the present work is the proposal of an indicator that relates flexibility with controllability; this indicator being the condition number. It is important to emphasize that there is no previously published work that relates condition number with flexibility. With the early evaluation of the condition number in the design of industrial processes, green chemistry processes along with a green engineering approach will guarantee sustainable processes. Furthermore, it is evident that by having a single indicator such as condition number, it will make it easier to implement in optimization processes, thus resulting in cheaper processes, and ensuring compliance with environmental and safety indicators. The study of the condition number as a quantitative measure of process flexibility was carried out on three different solar silicon production processes, which are described in the following Methodology section.

2. Methodology

The objective of this work aims to find a quantitative parameter, using the condition number to simultaneously measure the controllability and flexibility of the processes with the purpose of generating a simpler measurement.

The condition number and the minimum singular value provide important information in the theoretical control for a given frequency in a transfer function matrix within a multivariable system. It is the relationship between the maximum and singular value and represents the sensitivity of the system to uncertainty presented in the input. Therefore, it is possible to have the condition number [15] as a reference to measure the flexibility of a process [16].

The methodology to calculate the controllability through the condition number is different to the methodology to calculate the flexibility through the Resilience Index. The results obtained from these calculations cannot be mathematically related since the condition number is calculated in a state dynamic and represents a dimensionless value, while flexibility is calculated at a steady state and represents a percentage value. Despite these limitations, the relationship between controllability and flexibility through the condition number is empirically possible.

2.1. Condition number

The condition number (γ^*) can be specified as the quotient between the maximum singular value and the minimum singular value, as shown in Eq. (1).

$$\gamma^* = \frac{\sigma^*}{\sigma_*} \quad (1)$$

The magnitude of the minimum singular (σ_*) value quantifies the minimum distance to the nearest singular matrix. Being also a measure of the invertibility of the system, it indicates how fast the set point changes. The magnitude of the maximum singular (σ^*) value exhibits the potential problems that the system would present when implementing a feedback control scheme, indicating how quickly the disturbance is eliminated [24].

The condition number (γ^*) measures the sensitivity of the system to process parameter inaccuracies and mode errors; therefore, it is necessary to identify systems with minimum values of condition numbers and maximum values of the minimum singular value. Once this is done, it is expected that these systems will have the best dynamic behavior.

The quantitative version of the condition number is considerate using the strategy proposed by Cabrera et al. [16], where the area under the curve of the condition numbers with the addition of the singularities of its curve is calculated. The area value is performed for different frequency values (ω<=1, ω<=100, ω<=1000 rad/s), due that previously Morari and Zafiriou [25] establish that some frequencies lack of physical meaning. Then the evaluation of the area according to Cabrera et al. [16] occurs as show Eq. (2).

$$A_\gamma + \gamma_{sm} \quad (2)$$

Where

$$A_{\gamma_{b+3}} = \frac{\int_{-4}^{i} \log(\gamma) d\tilde{\omega}}{b-4} \quad (3)$$

$$\tilde{\omega} = \log\omega \quad (4)$$

b is the upper limit to evaluate

$$\gamma_i' = \frac{\log(\gamma(\omega_i)) - \log(\gamma(\omega_{i-1}))}{\log(\omega_i) - \log(\omega_{i-1})} \quad (5)$$

$$|\Delta\gamma_i'| = |\gamma'_{i+1} - \gamma'_i| \quad (6)$$

$$\gamma_{sm_i} = \Delta\log(\omega)|\Delta\gamma_i'| \quad (7)$$

2.2. Flexibility

Flexibility index was executed using Eqs. (1) and (2), corresponding to inscribing the largest proposed polytope within the feasible region. The flexibility index is equal to the distance between the vertex (V) and the nominal operating point (0) [20].

$$RI = \min_i \left\{ \left| l_i^{max} \right| \right\} \quad (8)$$

$$s.t. \left\{ \max_j f_j(\theta) \le 0 \forall l : \sum_i |l_i| \le RI \right\} \quad (9)$$

Based on a systematic procedure involving the following basic steps:

Identification of active sets, responsible for the feasible operation of a given design d and are therefore, a potential limitation of the process flexibility. They showed that, in certain conditions, each set has n+1 active restrictions and satisfies the following system of linear equations:

$$\sum_{j \epsilon J_A^k} \lambda_j^k = 1 \quad (10)$$

$$\sum_{j \epsilon J_A^k} \lambda_j^k \frac{\partial f_j}{\partial z} = 0, \quad \lambda_j^k \ge 0, \ j \epsilon J_A^k \quad (11)$$

where λ_j^k are constants, and J_A^k are the sets of the $n+1$ restrictions involved in the k[th] active set, k=1, …, n_{AS}. The n_{AS} set can be identified by analyzing Eqs. (4) and (5) with the signs of the gradients $\frac{\partial f_j}{\partial z}$ since these are constants for the linear case.

For each active set J_A^k a feasible function $\psi^k(d,\theta)$ of design d and parameter value θ qualitatively represents the adjustments of control variable z to minimize the maximum violation of the restrictions, defined as follows:

$$\psi^k(d,\theta) = \min_{z,\ u} u \quad s.t. \ f_j(d,\ z,\ \theta) \le u, \ j \epsilon J_A^k \quad (12)$$

Where u is scalar. If $\psi^k(d,\theta) \le 0$ for every value o θ and all active sets J_A^k $k = 1, \ \ldots, n_{AS}$, then the feasibility of operation can be ensured for design d. If $\psi^k(d,\theta) > 0$ for some values of θ, the design d is infeasible [26].

2.3. Case studies

Five processes were performed on the commercial scales using ASPEN Plus V8.8: three of them based in the work of Ramírez-Márquez et al. [27], were for the production of solar grade silicon (0.99999 mole fraction); Siemens, Union Carbide and Hybrid (a union between Siemens and Union Carbide), where a stochastic based optimization to decide on the column design and on the operating conditions for each process was achieved with three objectives in mind: economic objectives, using Return on Investment (ROI), safety objectives, applying an Individual Risk (IR), and environmental objectives, measured with Eco-indicator 99 (Eco99).

Siemens process, represented in Fig. 1, where metallurgical grade silicon is obtained by the reduction of quartz (SiO_2) with carbon. In Fig. 2, the results of the optimization with the sequences selected for purposes of this study, of Siemens process is shown. From Fig. 2, solutions have been selected considering sequences of the optimization with the best and worst results in optimization.

Union Carbide process, presented in Fig. 3 has a reactive distillation where the disproportionation of trichlorosilane is made. This process uses silane as a source of silicon, with the results of the optimization with the selected sequences to carry out this study is represented in Fig. 4, the solutions have been selected considering sequences of the optimization with the best and worst results in optimization.

The Hybrid Process between the Siemens and Union Carbide processes, represented in Fig. 5, also obtains metallurgical grade silicon by carboreduction of SiO_2, with its respective results of the optimization with the sequences selected to carry out this study are represented in Fig. 6, the solutions have been selected considering sequences of the optimization with the best and worst results in optimization.

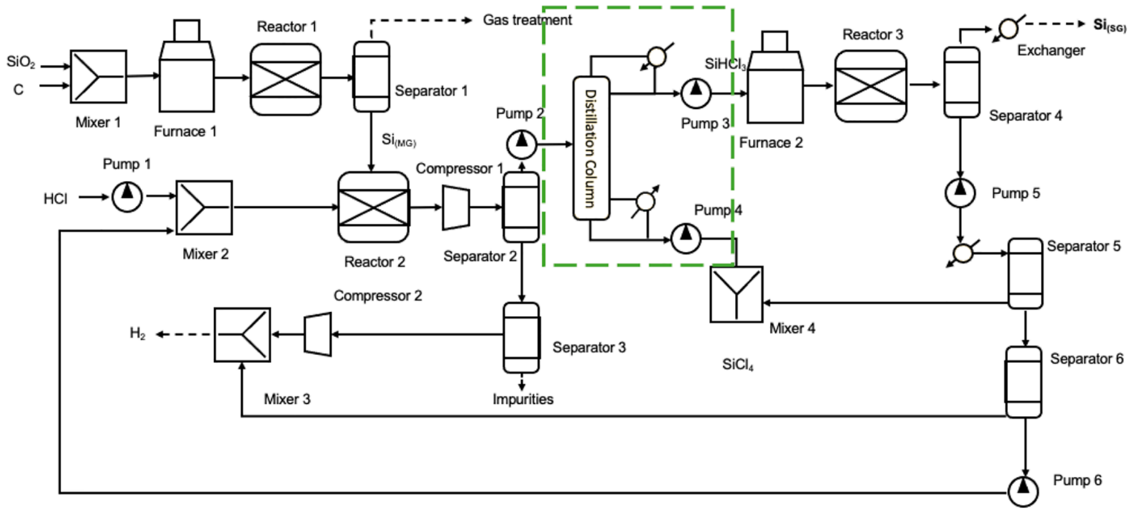

Fig. 1. Flowsheet of the Siemens Process.

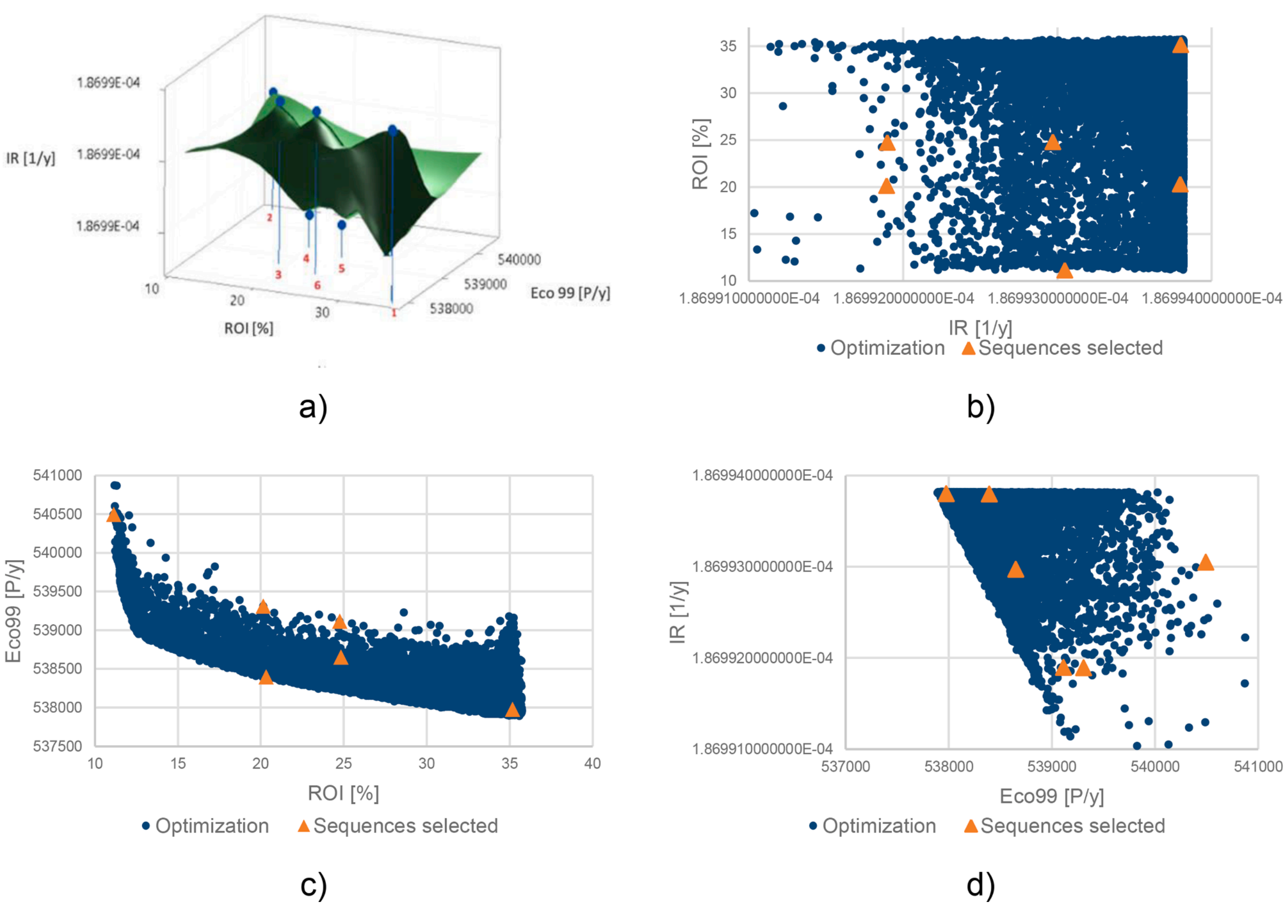

Fig. 2. Results of the optimization of the Siemens Process.

Additionally, a Binary distillation, is presented in Fig. 7 that separates 50% pentane and 50% hexane (0.98 mole fraction) and intensified Petlyuk scheme, is presented in Fig. 8, to separate propane-butane-pentane (0.99 mole fraction in dome and bottom, 0.98 mole fraction in side stream). All the sequences presented were obtained considering the complete set of mass and energy balances, equilibrium relationships, and summation constraints along with the phase equilibrium calculations.

For the study of the condition number as a quantitative measure of the flexibility, only the distillation columns of Siemens, Union Carbide and Hybrid processes were considered.

After optimization, the study of the flexibility and condition number was carried out in six representative sequences in five processes.

2.4. Flexibility assessment

To evaluate the flexibility of a process, the process conditions must be analyzed and for each control variable determined:

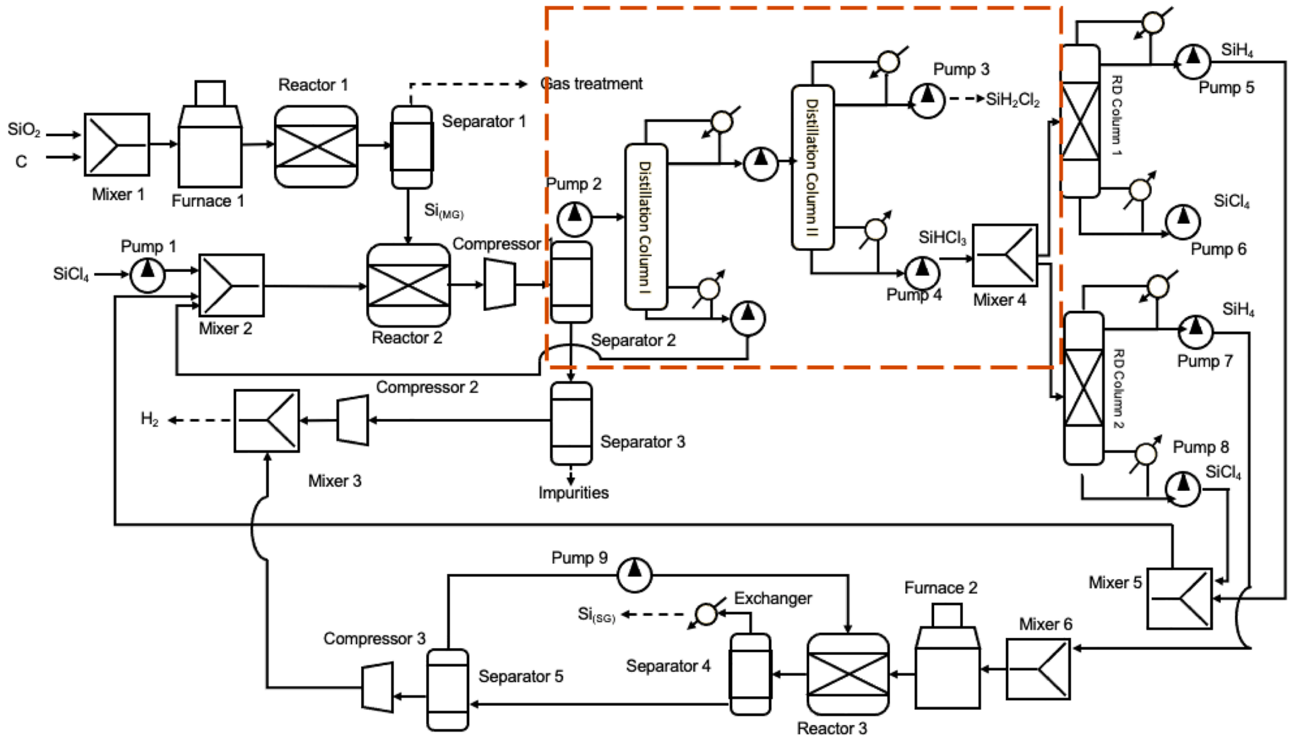

Fig. 3. Flowsheet of the Union Carbide Process.

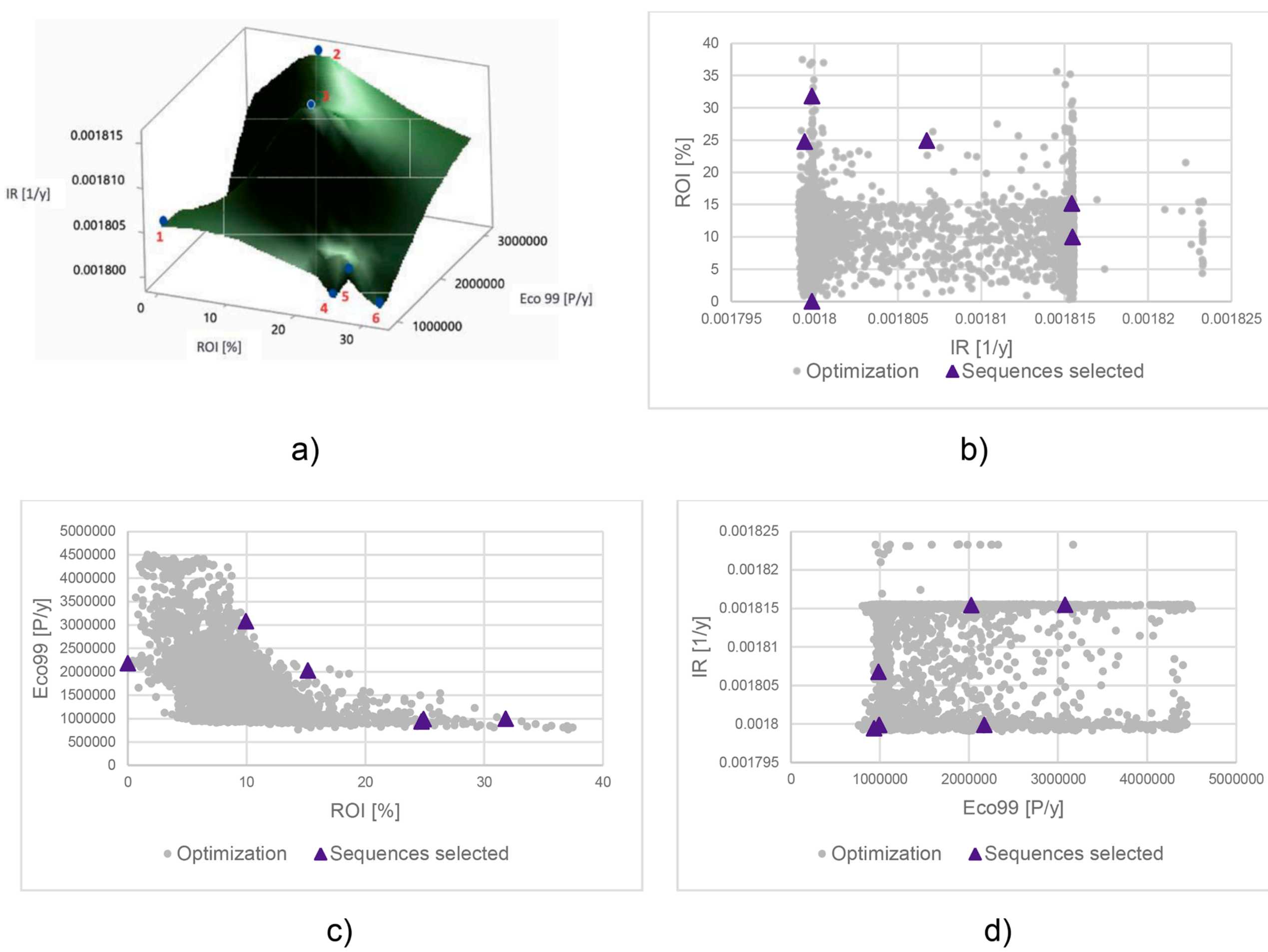

Fig. 4. Results of the optimization of the Union Carbide Process.

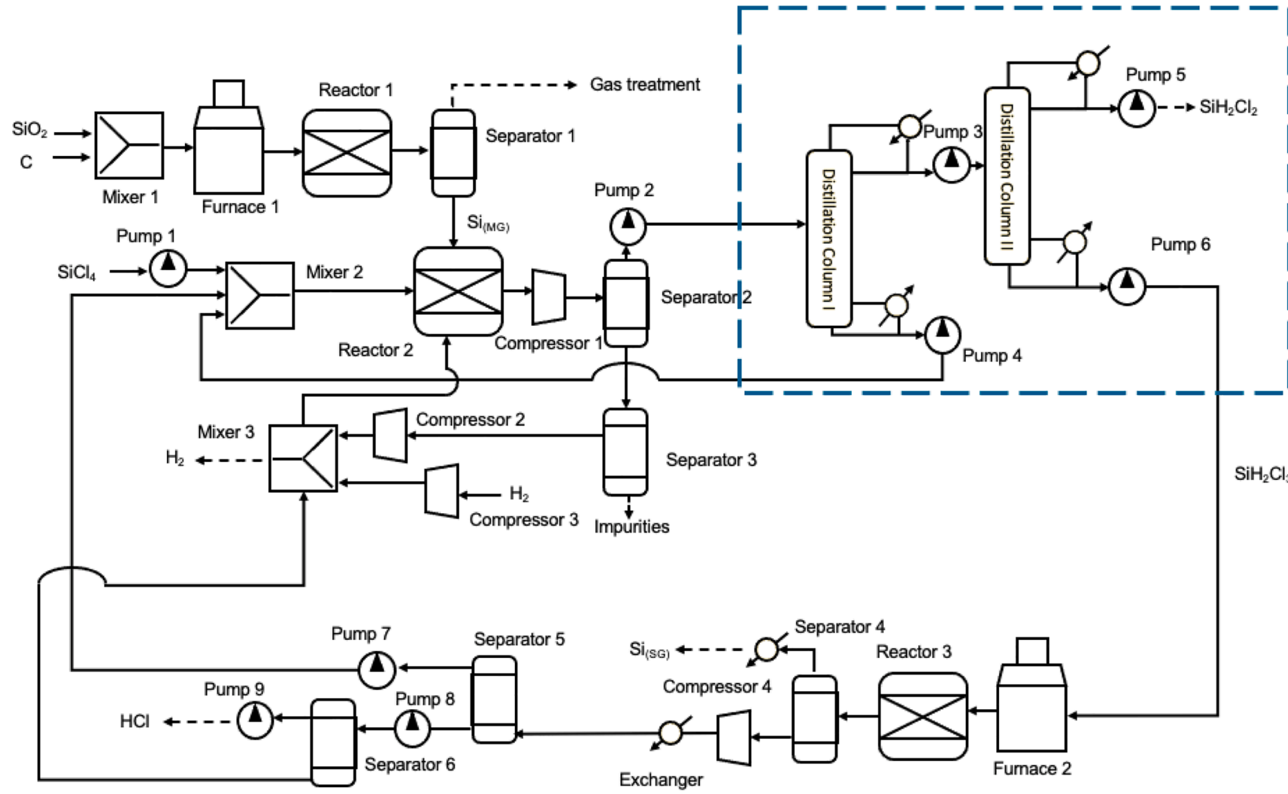

Fig. 5. Flowsheet of Hybrid Process.

(i) Lower and upper boundaries for the associated flexibility.
(ii) Estimate of the associated flexibility using the numerical sensitivity technique.
(iii) Determination of the value of flexibility by its limits, using the information obtained and the notion of independence.

To make a comparison between flexibility and the condition number, the values of the flexibility were normalized with respect to the maximum value of each range to consequently being able to compare both magnitudes to determine the relationship between them (see Eq. (13)), where the scheme with the maximum flexibility will have a value of 1.

$$Flexibility\ Index_{max} = \frac{Flexibility\ index}{Flexibility\ Index_{max}} \tag{13}$$

Fig. 9 shows the methodology for calculating the flexibility index.

2.5. Condition number assessment

To associate flexibility with a quantitative value of the controllability, the condition number of each sequence is calculated using the methodology [16] presented below:

(a) A disturbance of 1% was applied to each of the sequences in the reflux and reboiler duty below its initial value to study the dynamic behavior in the Aspen Dynamics simulator.
(b) Extraction of the open loopś responses.
(c) Adjustments in the Excelś software to the transfer functions models, estimating process gain (k), timeś constants (τ) and in some cases dead time (θ_d).
(d) Generation of the functionś matrix in the frequency's domain, according to the distillation columns and the exits that the studied processes have to realize the Singular Value Decomposition in MATLAB.
(e) The condition number $A_\gamma + \gamma_{sm}$ was evaluated from condition number at different frequency ranges (ω<=1, ω<=100, ω<=1000 rad/s).
(f) Construction of the graphics to relate the obtained values of flexibility and condition number. Fig. 10 shows the methodology for calculating the condition number.

Once the flexibility index and the condition number have been calculated, it will be possible to see their trend and decipher if there is a guideline for both, in complex and in simple processes. In order to make a comparison between the condition number singularity convolution ($A_\gamma + \gamma_{sm}$) (dimensionless value) and the flexibility (percentage value), the values of the convolution were normalized with respect to the maximum value of each range to be able to express them as a percentage and consequently being able to compare both magnitudes to determine the relationship between them (see Eq. (14)), where the scheme with the maximum convolution will have a value of 100%.

$$|A_\gamma + \gamma_{sm}|_{max} = \frac{(A_\gamma + \gamma_{sm})_i}{(A_\gamma + \gamma_{sm})_{max}} \times 100 \tag{14}$$

3. Results

This section presents the results of the flexibility and ($A_\gamma + \gamma_{sm}$) values calculated under the methodologies shown above. Three solar grade silicon processes, a binary distillation column, and a Petlyuk column were evaluated. For the first three solar silicon processes, the values obtained for Return on Investment (ROI), Eco-indicator 99 (Eco99), and Individual Risk (IR) were retrieved from the work of Ramirez-Marquez et al. [27]. By rescuing these values, an analysis of sustainability with respect to the number of conditions as an indicator of flexibility can be made. The results will be divided by each of the processes studied: Siemens Process, Union Carbide Process, Hybrid Process, Binary Distillation Column, and Petlyuk Column. Six different sequences

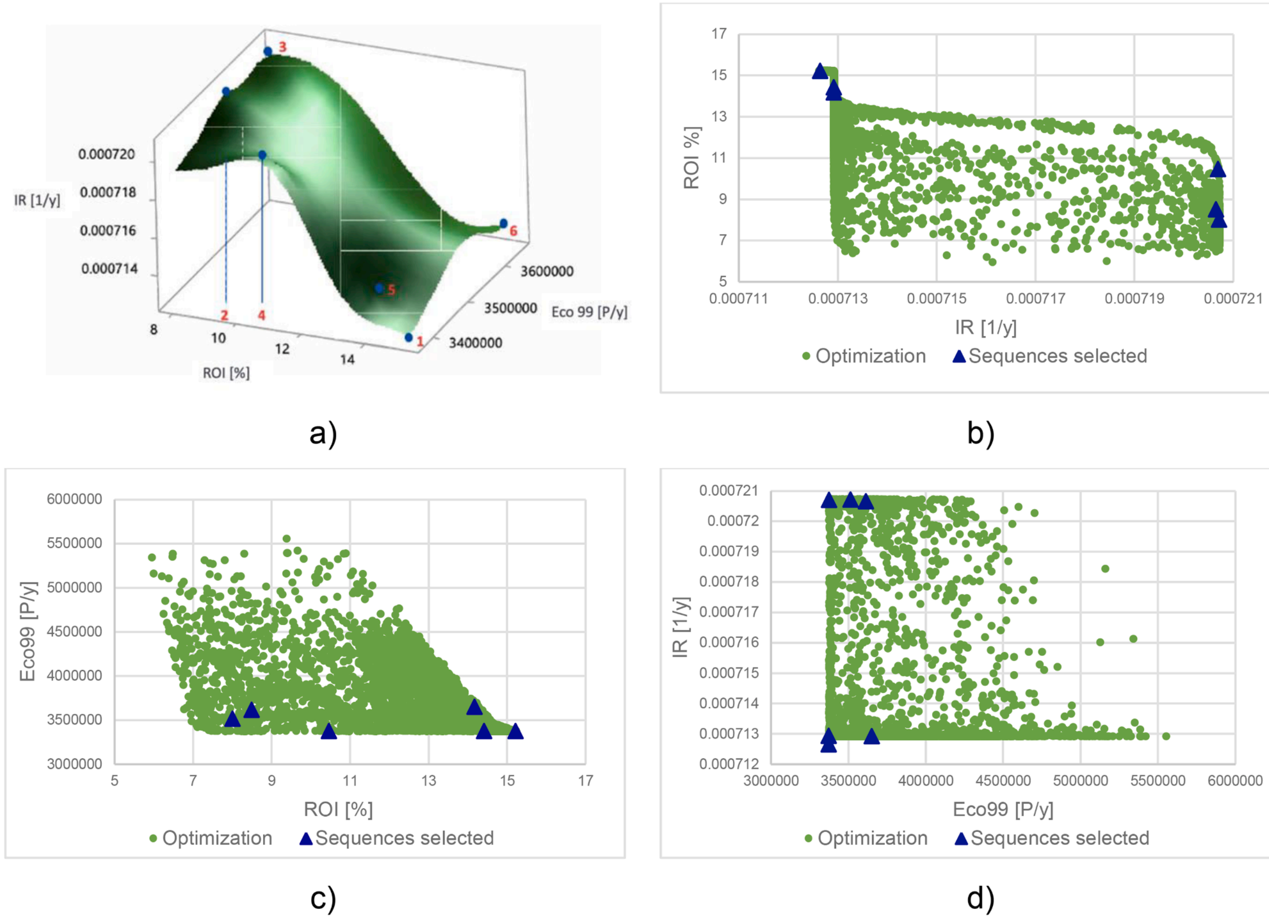

Fig. 6. Results of the optimization of the Hybrid Process.

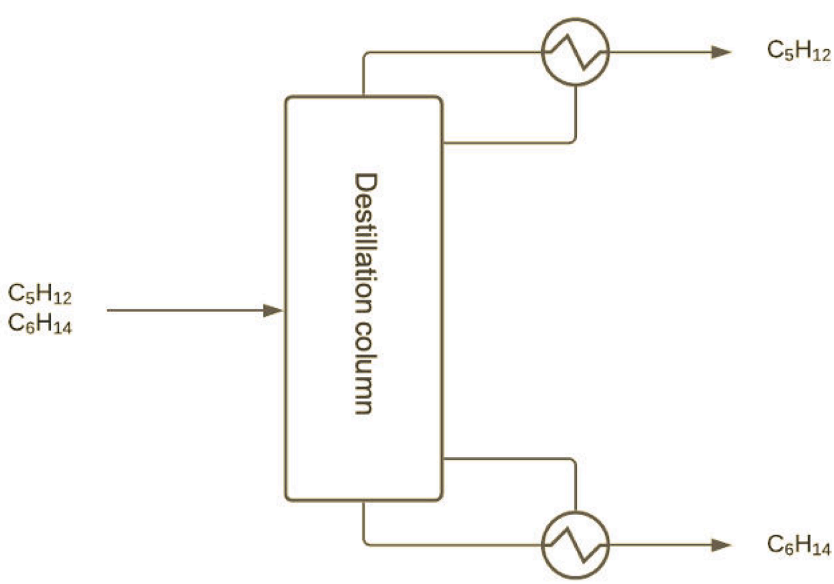

Fig. 7. Flowsheet of the binary distillation column.

of each process were evaluated.

Since the flexibility index is calculated at low frequencies, the ($A_\gamma + \gamma_{sm}$) with frequencies less than or equal to 1 is the most appropriate one to compare to the flexibility index, since it is the one that can be best associated with the trend sought. Figs. 12, 15, 18, 19 and 20, present the flexibility index with the respective ($A_\gamma + \gamma_{sm}$) with ω<=1 for each of the analyzed sequences.

The analysis of the condition number, with frequencies less than or equal to 1, in each of the analyzed sequences follows the trend that was originally intended, the ($A_\gamma + \gamma_{sm}$) behaves in the opposite way to flexibility. In other words, when flexibility is high, the condition number tends to be low.

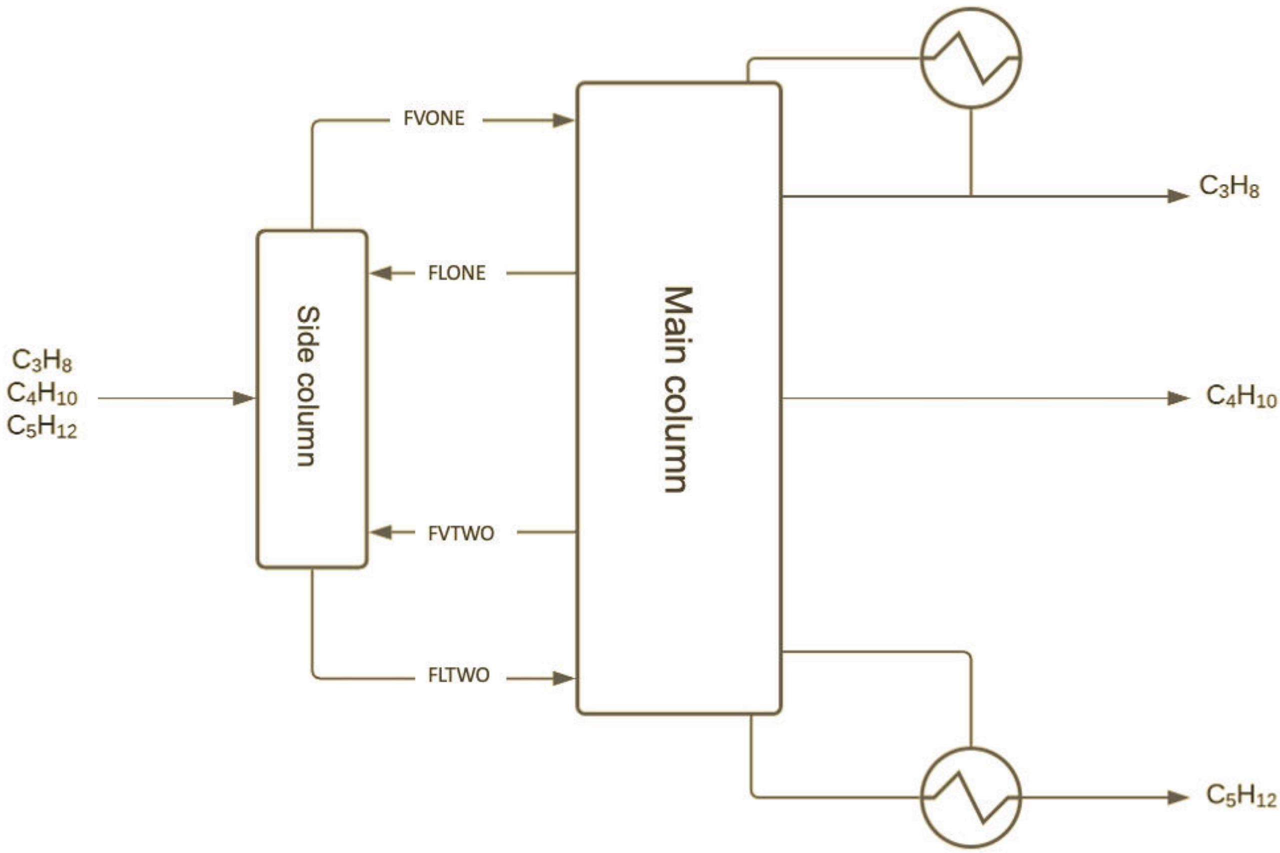

Fig. 8. Flowsheet of the Petlyuk distillation Column.

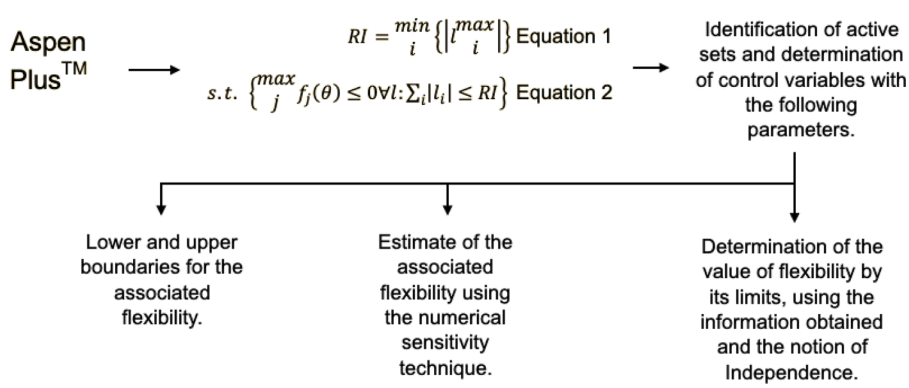

Fig. 9. Methodology to calculate the flexibility index.

Having obtained all the information and after observing the opposite trend of the $(A_\gamma + \gamma_{sm})$ with respect to the flexibility index, it can potentially be said that the $(A_\gamma + \gamma_{sm})$ is an indicator that can simultaneously measure controllability and flexibility.

3.1. Siemens process results

In Fig. 12, it can be observed that the behavior of the $(A_\gamma + \gamma_{sm})$ at high frequencies (ω<=1000, ω<=100) tends to present values close to those of the flexibility index; this being a different behavior than expected, unlike $(A_\gamma + \gamma_{sm})$, where a low frequency (ω<=1) is presented, and the desired trend can be observed.

Analyzing the sequences in Figs. 11 and 12.

Sequences 1: has a low $(A_\gamma + \gamma_{sm})$ and a low flexibility index. Within all the sequences, it is the point that least presents the desired trend. It is also the column with the smallest diameter and has few stages, which explains why the flexibility index is low, even presenting the best ROI (35.171%) and the best Eco99 (537975.844 P/y). The IR is the highest (1.869937993804E-04 1/y), which makes it a sequence compared to the others, good in terms of controllability, profitability, and sustainability, but bad in flexibility and safety, making it unfeasible (see Fig. 11).

Sequence 2: tends to be more flexible because the dimensions of the equipment are larger. Reviewing Fig. 11 where the dimensions of the Siemens process are registered, with each of the distillation columns at each point, it is observed that although the diameter is small compared to the others (0.8906 m), the height of the equipment and the number of stages is high; this makes it easier to absorb the disturbances that may occur, making it a flexible sequence with a low $(A_\gamma + \gamma_{sm})$. This is an indicator that the system in sequence 2 passing from one stationary point to another was stabilized in a short time and did not present many

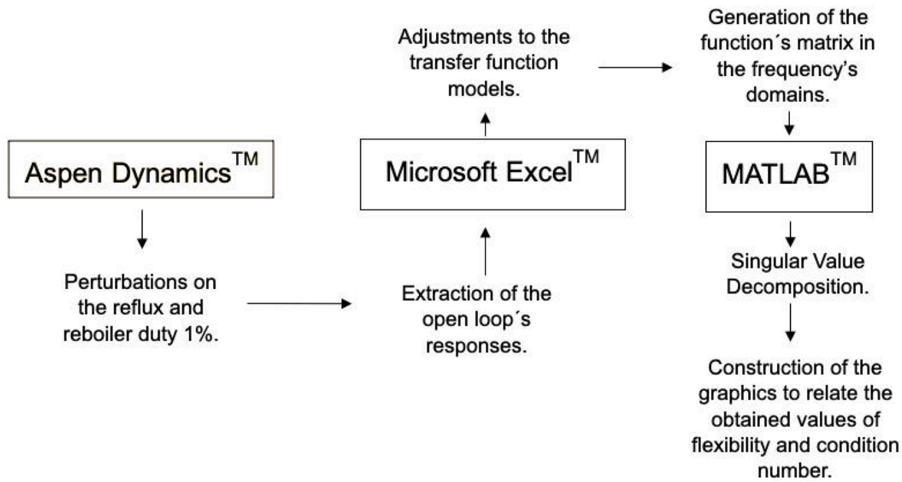

Fig. 10. Methodology to calculate condition number.

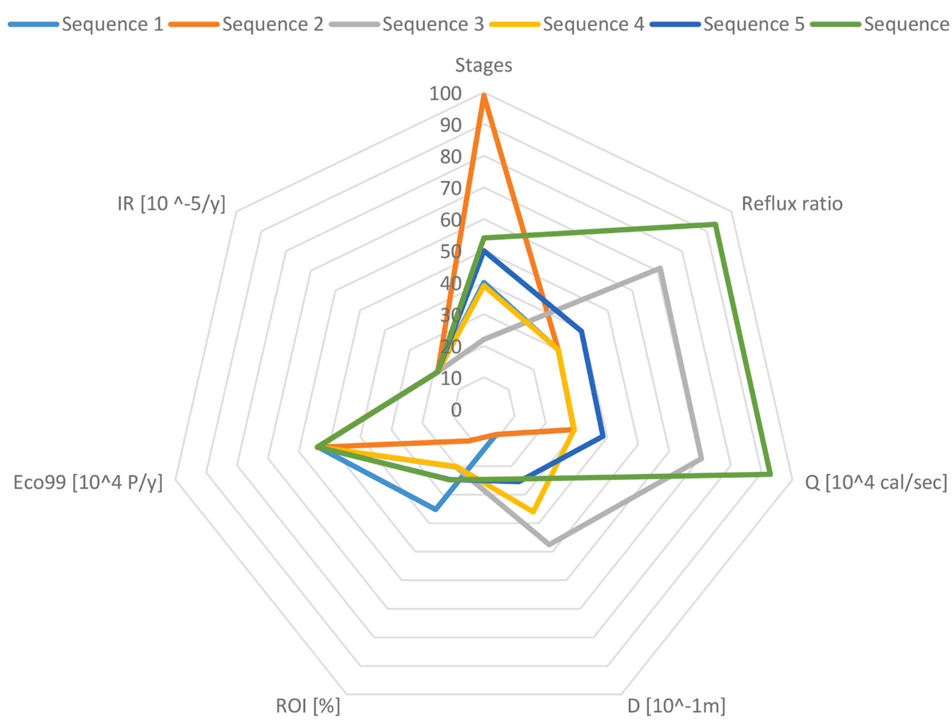

Fig. 11. Topology of the separation sequences in the Siemens Process.

oscillations.

Sequence 3: has the best $(A_\gamma + \gamma_{sm})$, yet one of the lowest flexibility indexes. The low flexibility in the sequence is because it is the column with the fewest stages, even though it has the largest diameter. Fig. 11 shows that the IR (1.869937961717E-04 1/y) and ROI (20.319%) are low, along with a high value for the Eco99 indicator (538395.593 P/y). Making the sequence unprofitable, environmentally damaging, and inflexible.

Sequence 4: presents one of the highest $(A_\gamma + \gamma_{sm})$ and the least flexibility of all the sequences, even though it has a large diameter. The height and the number of stages is low, and any disturbance to the system will damage it significantly; and though it presents the best risk index of the six sequences analyzed, the process is not profitable under these conditions.

Sequence 5: is the one with the best flexibility, together with the highest value of controllability, the column has a high number of stages and a high diameter, which explains why the flexibility is good. It also helps to present a good ROI (24.756%) but presents a high Echo indicator 99 (539115.916 P/y). Even with this data the ROI is low and represents one of the safest sequences of all those analyzed in the Siemens Process.

Sequence 6: presents the highest flexibility index and one of the lowest $(A_\gamma + \gamma_{sm})$, based on the data in Fig. 11, this sequence has a large diameter (2.4480 m) and a high number of stages (54 stages), which ensures that there is little change from one steady state to another the sequence will stabilizes quickly.

3.2. Union carbide process results

In Fig. 15, it can be observed that the behavior of the $(A_\gamma + \gamma_{sm})$ at high frequencies (ω<=1000, ω<=100) tends to present values close to those of the flexibility index; a behavior different to that expected

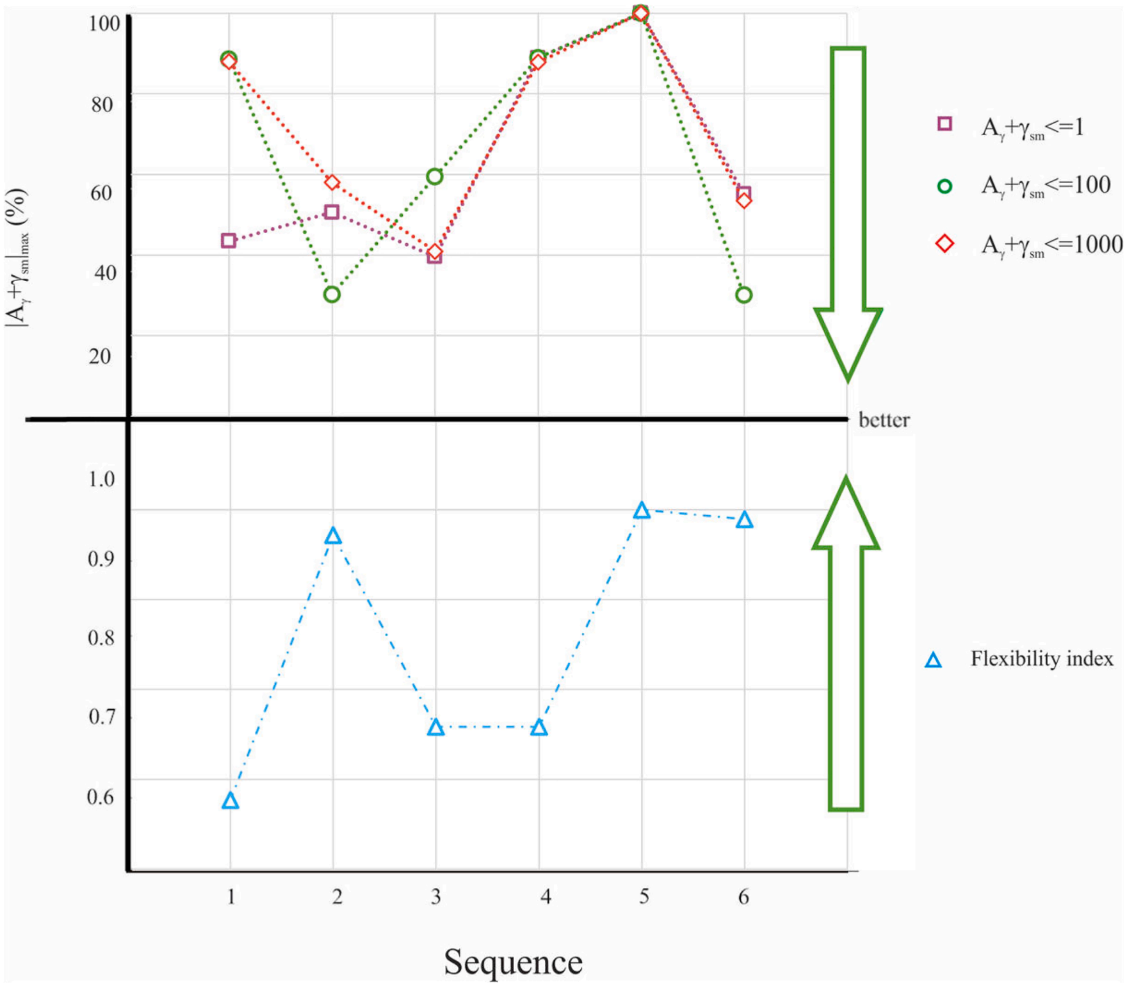

Fig. 12. Flexibility and condition number plot at different frequencies (ω<=1000, ω<=100, and ω<=1) in representative sequences analyzed from the Siemens Process.

(ω<=1), where it is observed that at low frequencies and in most the sequences. If the flexibility is good, the control is also good.

Analyzing the sequences in Figs. 13–15.

Sequence 1: presents the lowest flexibility index and the highest ($A_\gamma + \gamma_{sm}$). After reviewing Figs. 13 and 14, where the dimensions of the first two columns proposed for the Union Carbide process are found: sequence 1, with the largest diameters of all the sequences (Column 1-3.9108 m, and Column 2-4.8352 m), but with the lowest number of stages and the lowest ROI (0.024%). This sequence is also not profitable (Figs. 13 and 14).

Sequence 2: has a high flexibility, the ($A_\gamma + \gamma_{sm}$) are low, and there is a notable improvement. Observing Figs. 13 and 14, it has a large diameter (Column 1-1.5296 m, and Column 2-1.9337 m) and a high number of stages (55 and 32 stages), which ensures that the change from one steady state to another does not cost great work to the system.

Sequence 3: shows a flexible system, with a high flexibility index and controllability with a low ($A_\gamma + \gamma_{sm}$). This can be verified by looking at the dimensions of the design, since it has a high number of stages (96 and 40 stages) and a large diameter in the first column (2.1383 m) Even with good attributes in its controllability and flexibility, the sequence is not economically viable, with a high IR (0.00181543 1/y), making it unsafe (Fig. 13).

Sequence 4: has a good flexibility index and a low ($A_\gamma + \gamma_{sm}$), indicating a good control. It is a sequence whose design has a high number of stages in its columns, but small diameters. Nevertheless, the height derived by the high number of stages will help the process with stability and flexibility. Even with a good ROI (24.743%) and IR (0.00179943 1/y), the Eco indicator 99 (935856.9598 P/y) is high, which does not make it quite compatible with the metrics and green chemistry (See Figs. 13 and 14).

Sequence 5: exhibits a low flexibility index and a high ($A_\gamma + \gamma_{sm}$), making it a poor sequence. This can also be checked when reviewing the dimensions of the design as it has a low number of stages, and a high IR (0.00180674 1/y). This is consistent with poor controllability and control. The attributes of this sequence are a high ROI (24.906%) and a high Eco99 indicator (987173.6568 P/y).

Sequence 6: has one of the highest flexibility indexes and the lowest ($A_\gamma + \gamma_{sm}$) of all the presented sequences of the process. It has large diameters (Column 1-1.6035 m, and Column 2-1.1897 m), and high numbers of stages (61 and 42 stages), which gives veracity to the behavior of the flexibility and the ($A_\gamma + \gamma_{sm}$).

3.3. Hybrid process results

In Fig. 18, it can be observed that the behavior of the ($A_\gamma + \gamma_{sm}$) at high frequencies (ω<=1000, ω<=100) tends to present values different from those expected. And just like in the Siemens Process and Union Carbide, the ($A_\gamma + \gamma_{sm}$) at low frequency (ω<=1) is the most adequate to the sought trend.

Analyzing the sequences of Figs. 16–18:

Sequence 1: presents a high flexibility and a low ($A_\gamma + \gamma_{sm}$), analyzing Figs. 16 and 17, it is a sequence that presents large diameters (1 m each one) and a low number of stages (47 and 81 stages) compared

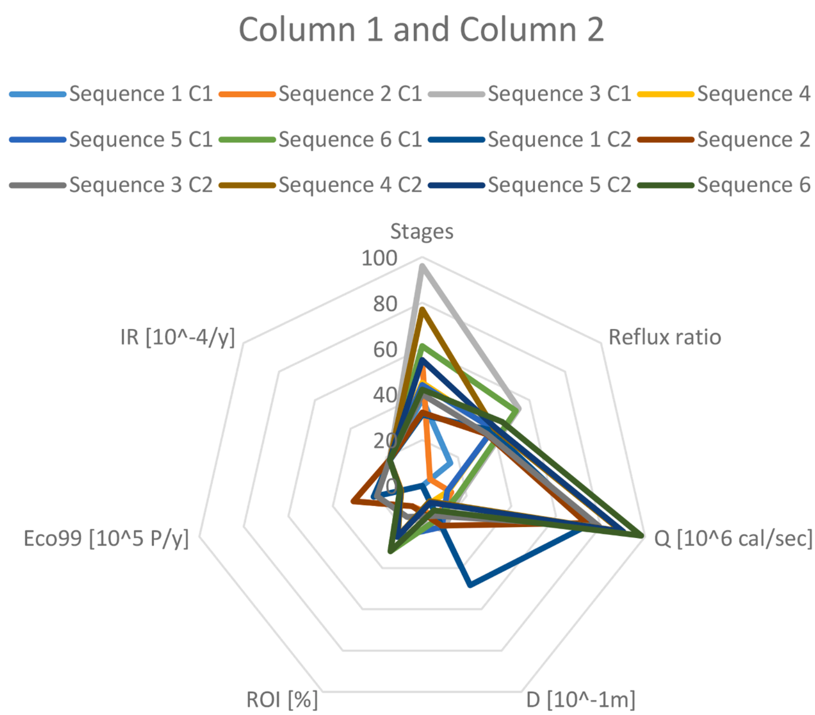

Fig. 13. Topology of the distillation columns in the Union Carbide Process.

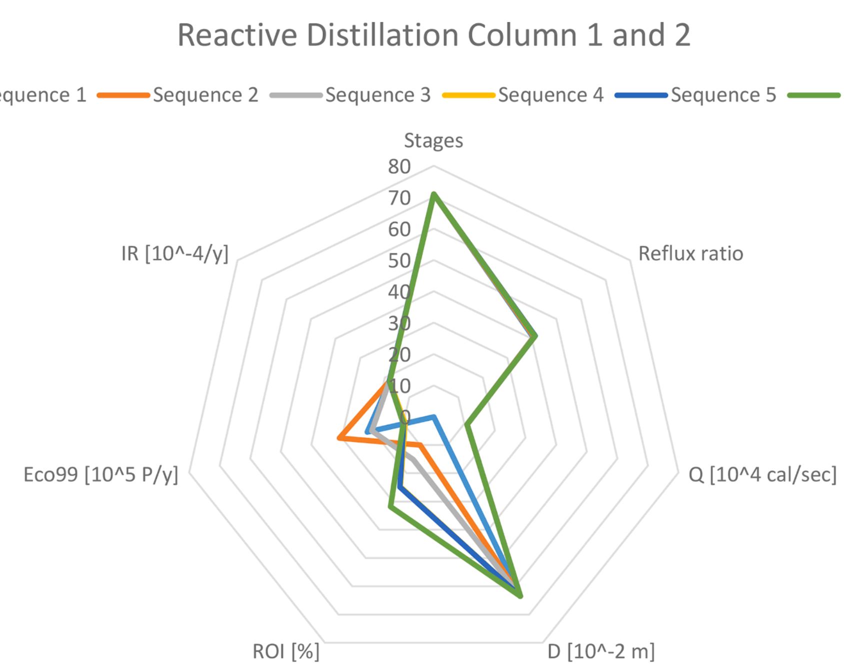

Fig. 14. Topology of the Reactive distillation columns in the Union Carbide Process.

to the others. This confirms that, due to the large dimensions derived from the diameter, the sequence presents a good behavior both in flexibility and in the ($A_\gamma + \gamma_{sm}$).

Sequence 2: exhibits a high ($A_\gamma + \gamma_{sm}$) and a low flexibility index, even with a large diameter (Column 1-3.2988 m, and Column 2-1.9764 m) and many stages (95 and 101 stages), it is a sequence in which, if disturbances occur, it will not have the ability to stabilize. It is the least expensive sequence of all, with a bad Eco99 indicator (3515046.15 P/y) and a high IR (0.000720714 1/y). With all these set of disadvantages, it is one of the most unfeasible sequences.

Sequence 3: is one of the most notorious sequences where the flexibility is low and the ($A_\gamma + \gamma_{sm}$) are one of the highest presented. By having a low number of stages (57 and 95 stages) in the two columns and one of the lowest diameters (Column 2-0.9839 m). It can be observed that it is one of the least profitable sequences, with the highest risk index (See Fig. 6).

Sequence 4: presents a high ($A_\gamma + \gamma_{sm}$) and a low flexibility. Analyzing Figs. 16 and 17, it can be observed that there is a small and a large diameter. Despite the large diameter of the first column, the sequence presents difficulty to absorb the disturbances. This indicates that the sequence does not have a good control. It also presents a high safety index and a low Echo indicator 99 (See Fig. 16).

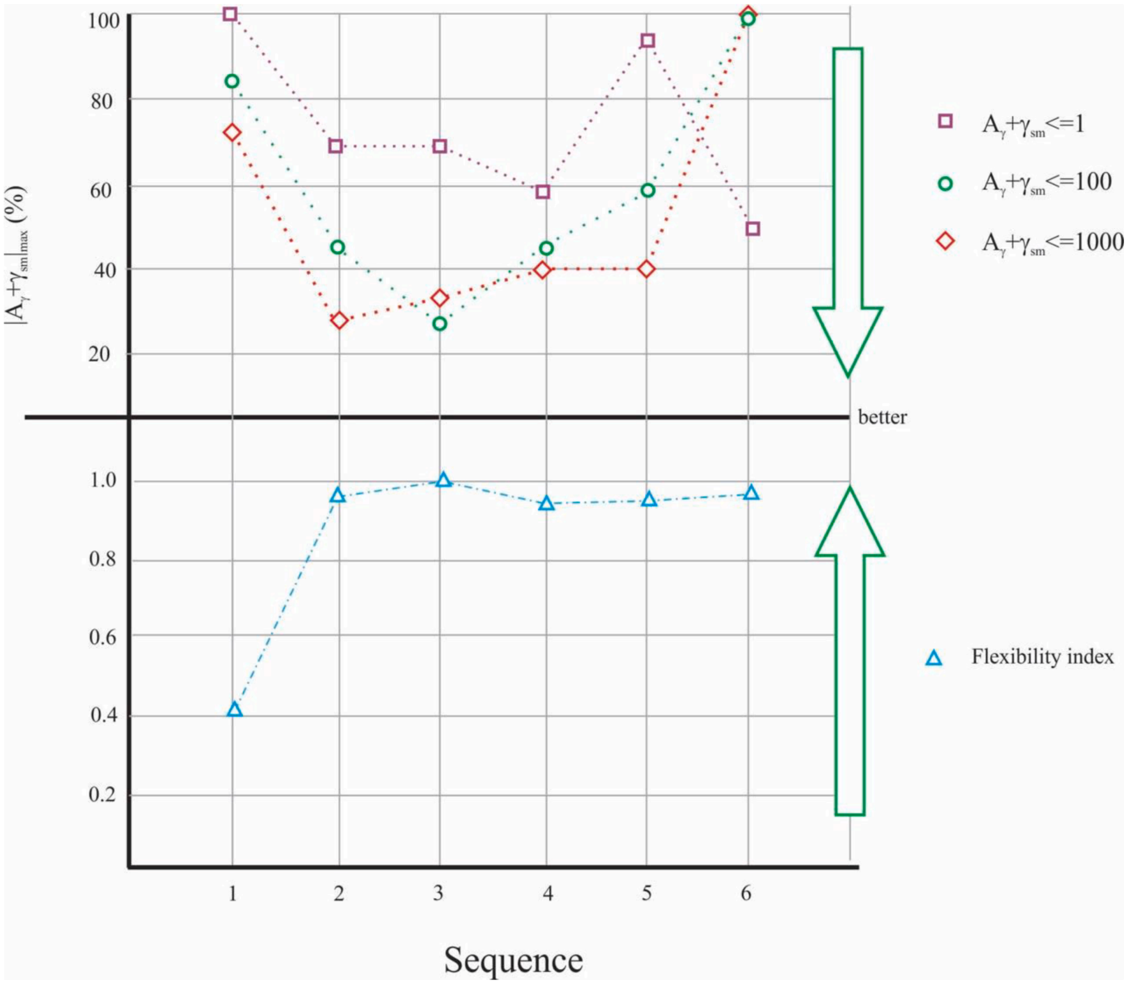

Fig. 15. Flexibility and condition number plot at different frequencies (ω<=1000, ω<=100, and ω<=1) in representative sequences analyzed from the Union Process.

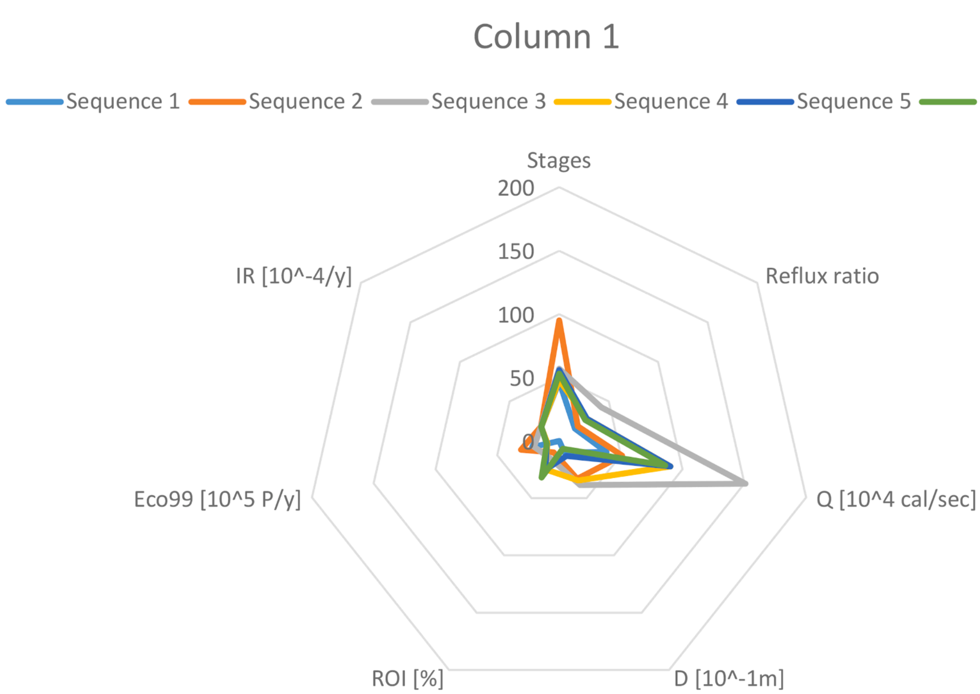

Fig. 16. Topology of the separation sequences in the Hybrid Process, Column 1.

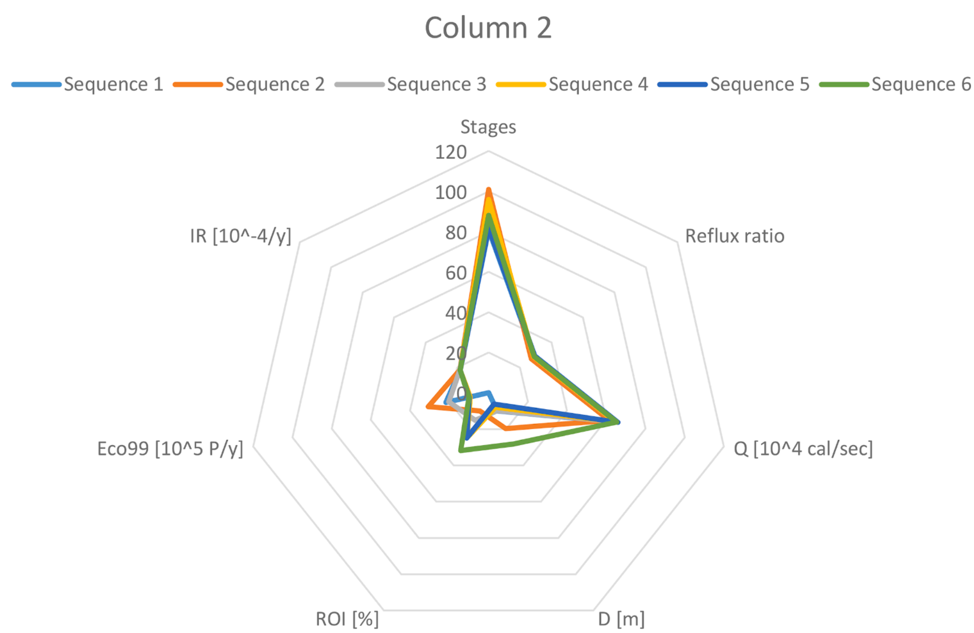

Fig. 17. Topology of the separation sequences in the Hybrid Process, Column 2.

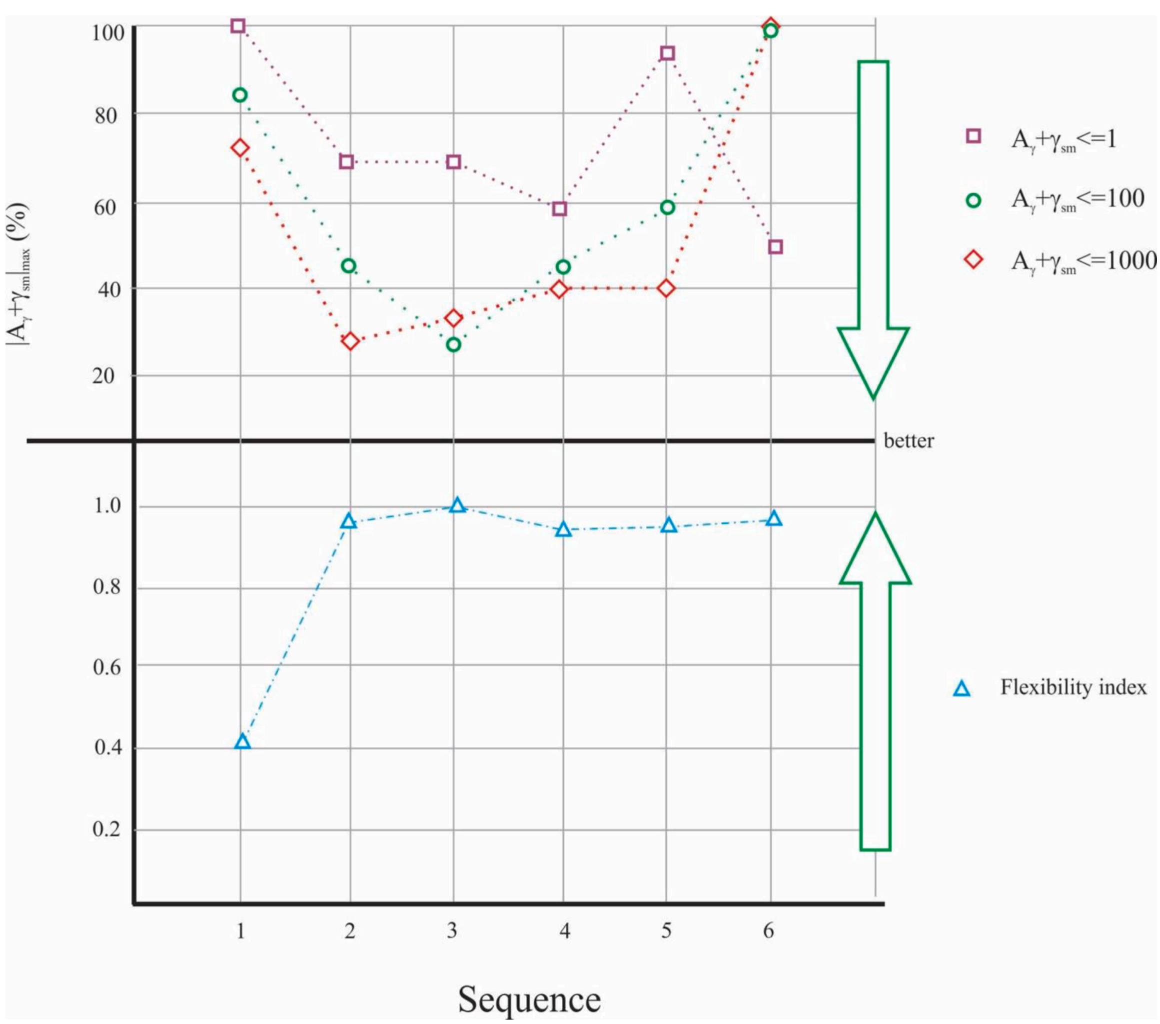

Fig. 18. Flexibility and ($A_\gamma + \gamma_{sm}$) plot at different frequencies (ω<=1000, ω<=100, and ω<=1) in representative sequences analyzed from the Hybrid Process.

Sequence 5: shows low controllability and flexibility since it has few stages and small diameters in the columns compared to the other sequences. Although it has a low IR (0.000712925 1/y), it has a high Eco99 (3374468.93 P/y), but a good ROI (14.419%). Even with a good ROI value, it is not a viable sequence since it there is bad data in its design, controllability, flexibility and in the optimization parameters (Figs. 16 and 17).

Sequence 6: has a high ($A_\gamma + \gamma_{sm}$) and a low flexibility index, which can be verified by observing the measurements presented in Fig. 16, column 1, where the stages and diameter dimensions are small (53 stages and 0.6746 m). This indicates poor control and flexibility. The advantages of this sequence are that it has one of the highest ROI (14.175%) and a good IR (Figs. 16 and 17).

3.4. Binary distillation column results

The study of the proposed indirect indicator was carried out in a binary distillation column (to separate a mixture in equal parts of pentane and hexane). The dimensions of the proposed sequences shown in Table 1 were also recorded. As in the previous case studies, the trend is more noticeable in the condition numbers calculated at frequencies less than or equal to one, related to a flexibility index calculated at low frequencies.

The case of a binary distillation column is, out of all the case studies, the one that presents the ideal behavior sought with this new indirect indicator.

As seen in Fig. 19, when controllability is good, with a low ($A_\gamma + \gamma_{sm}$), flexibility also tends to be good, with high flexibility index values, as well as large stage and diameter dimensions. On the contrary, when controllability is bad, indicated by a high ($A_\gamma + \gamma_{sm}$), flexibility tends to be bad, with low flexibility index values, as well as low stage and diameter values.

3.5. Petlyuk column results

As a last case study, the indicator was studied in a Petlyuk distillation column (to separate a mixture of propane, butane, and pentane). The trend in the ($A_\gamma + \gamma_{sm}$) calculated at frequencies less than or equal to one, related to a flexibility index calculated at low frequencies, can be seen in a simpler way. In this case, the stages, feed stage and Petlyuk column diameter were maintained (as shown in Table 2).

In the case of the Petlyuk distillation column, the graph presented in Fig. 20 shows that in sequences 1 and 6 there are a high ($A_\gamma + \gamma_{sm}$) related to a low flexibility index, and in sequences 2 and 4 there are a low ($A_\gamma + \gamma_{sm}$) and a high flexibility index. Even though not all the sequences present the expected tendency, the indirect indicator could be used to obtain good values of flexibility and controllability within the optimization, and toget her with these, it guarantees a safe process.

3.6. Analysis of the results

In most of the analyzed points of each sequence a certain tendency was observed. When the flexibility index has low values, the ($A_\gamma + \gamma_{sm}$) tends to go up, which means that both controllability and flexibility are bad; and just as when the flexibility index has high values, the ($A_\gamma + \gamma_{sm}$) tends to have low values, which represents good control and good flexibility in the system. It is here when it can be observed that the ($A_\gamma + \gamma_{sm}$), being the simplest to calculate, can indirectly indicate the behavior of flexibility.

In the specific case of the Siemens process it can be observed that the sequence with better control and flexibility are the ones with larger dimensions. If Fig. 11 is analyzed where the topology of the columns is shown, the better sequences tend to be the ones with larger dimensions, either the diameter or the number of stages. That is because while the columns are bigger, the control is going to be better as has been reported in the literature [28]. Being the contribution of this work and based in the results shown above, that also the flexibility is going to be preferable in a larger percentage of the cases when the dimensions of the columns are bigger, therefore, it allows to associate both concepts control, and flexibility as a single indicator. As shown in sequences 3, 4 and 5.

In the Union Carbide process the sequences that presents better results are 1, 4 and 6. As shown in Figs. 13 and 14 these sequences are the ones with a larger number of stages, and because of that the columns must be taller. Giving the process a better flexibility and control. In the case the Hybrid process also happens that the taller columns represented with the number of stages, as shown in Figs. 13 and 14, the sequences 2, 3, 4 and 6 are the ones that presents better control and flexibility results. In case of the binary distillation column, there is a 100% agreement between the condition number singularity convolution and the flexibility index, shown in the Table 1 and Fig. 19. Finally, in the case of the Petlyuk distillation column, the sequences 1 and 6 are the ones that presents the better tendency, and also according to the Table 2, have one of the highest reflux ratio.

The ($A_\gamma + \gamma_{sm}$), for these processes, near the steady state has an effect directly proportional to the behavior of flexibility, therefore, it indicates that there is a direct correlation between the ($A_\gamma + \gamma_{sm}$) and the flexibility. This opens the option that the condition number singularity convolution that can be used as an objective function of control and can simultaneously obtain a flexible optimum. This guarantees a sustainable and green process. One that can introduce three concepts in a large multi-objective optimization problem, and which can be used for future optimization work.

Another important point to highlight from these results, is that for the correlation to be used, it is necessary to work with low frequencies ($\omega \leq 1$) since high frequencies ($\omega \geq 1000$) change the behavior of the sequence. However, to work with low frequencies does not represent a drawback, since Cabrera-Ruiz et al., [16] state that a real process behaves at such frequencies ($\omega \leq 1$).

When, in a case study, there are concordances in the number of conditions at low frequencies ($\omega \leq 1$) and the flexibility index is between 50% to 80%, it can be assumed that the proposed index works.

In most processes it can be seen, with their respective scenarios, the agreement that the higher the flexibility index is, the lower the number of conditions will tend to be. Figs. 12, 15, 18–20, indicate two tendencies. One: that the condition number singularity convolution can be related to the flexibility and two: that there is a relation to the ability to remove disturbances from the system.

Thanks to the case studies mentioned, it can also be noted that the simpler the separation process is, the better the results of the indicator will be. As shown in the binary distillation column.

Table 1
Topology of the binary distillation column.

# Sequence	Stages	Feeding Stage	Reflux ratio	Q [cal/sec]	D [m]
1	17	9	1.525	267467.196	1.0024
2	80	34	1.137	239041.272	0.9375
3	65	31	1.135	237642.591	0.9389
4	28	13	1.155	236696.761	0.9516
5	37	22	1.133	235231.441	0.9461
6	44	26	1.132	235624.343	0.9442

4. Conclusions

This paper presents a quantitative measure to simultaneously evaluate the controllability of a process and its flexibility through the condition number singularity convolution. The results show that the condition number singularity convolution serves as a quantitative index of process controllability and that, in turn, this correlates with flexibility. Achieving a single indicator (condition number) that measures controllability and flexibility for the case studies presented. The most

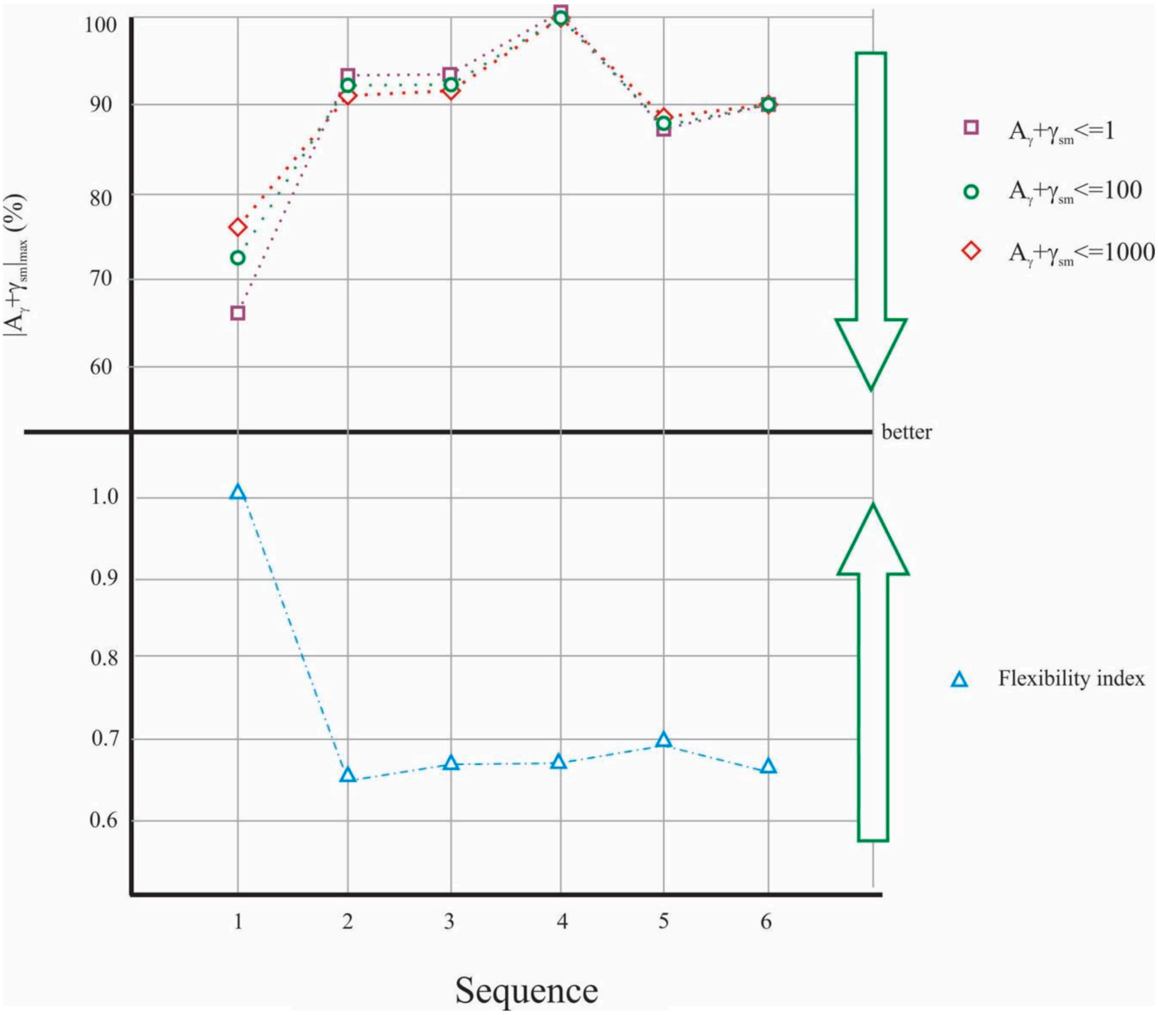

Fig. 19. Flexibility and ($A_\gamma + \gamma_{sm}$) plot at different frequencies (ω<=1000, ω<=100, and ω<=1) in representative sequences analyzed from the Binary Distillation Column.

Table 2
Topology of the petlyuk column.

# Sequence	Pre	Main Column Stages	Feeding Stage	Reflux ratio	Q [cal/sec]	D [m]
1	25	52	12	11.341	342216.460	1.106
2				9.993	307347.531	
3				6.450	216404.332	
4				5.639	195098.691	
5				6.147	207564.743	
6				7.100	232511.885	

controllable and flexible processes are the safest, fulfilling the sustainability criteria of Jimenez-Gonzalez and Constable [4]. And regardless of safety, in the cases exhibited, there is an evident relationship between better profitability and minimum environmental impact with the minimization of the condition number. Nevertheless, high frequencies have more consistent values (ω≥1000), at low frequencies (ω≤1) it could be a reliable index to measure both items. It is important to mention that low frequencies are usually present in the industry, i.e., disturbances that do not exceed 5% of the set-point or of the feed flow [28]. High frequencies represent disturbances with large percentages, which are not common in the industry.

All case studies are separation processes, specifically conventional and reactive distillation columns. Then having relatively large diameters in the design, and a high number of stages, becomes a necessary condition to associate controllability with flexibility simultaneously. Moreover, the relationship between both parameters became very clear for the simple processes and became less evident while process complexity increases.

With this in mind, and index could be provided to ensure that future optimizations have economic, affordable, green, and controllable processes while respecting and enhancing an objective that is currently of much importance within sustainable processes: safety. Considering the results obtained, the merging for the first time of the flexibility and controllability indicators into a single indicator, such as the number of conditions, seems to be an important advance in the field of green and sustainable process design.

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Conceptualization, Methodology, Validation, Results, Manuscript, Original Draft, Editing & Revising. **Ramírez-Márquez César:** Conceptualization, Methodology, Validation, Results, Manuscript, Original Draft Editing & Revising, Funding acquisition. **Cabrera-Ruiz Julián:** Conceptualization, Methodology, Validation, Results, Manuscript, Original Draft, Editing & Revising. **Segovia-Hernández Juan Gabriel:** Conceptualization, Methodology, Validation, Results, Manuscript, Original Draft, Editing & Revising, Funding acquisition.

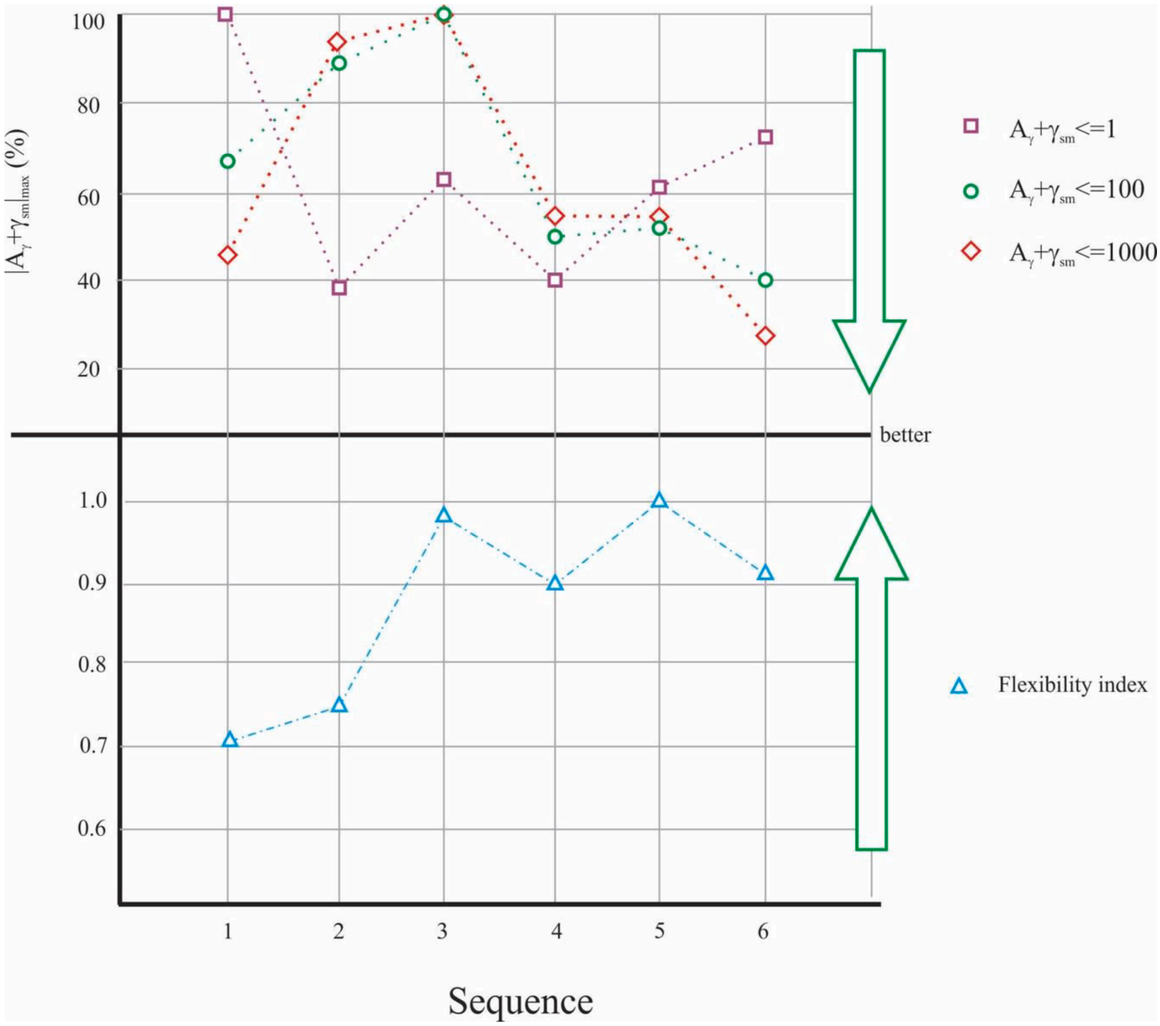

Fig. 20. Flexibility and ($A_\gamma + \gamma_{sm}$) plot at different frequencies (ω<=1000, ω<=100, and ω<=1) in representative sequences analyzed from the Petlyuk Column.

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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.

Data availability

The data that has been used is confidential.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.cep.2022.109184.

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