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Sustainable Process Design for Acetone Purification Produced via Dehydrogenation of 2-Propanol

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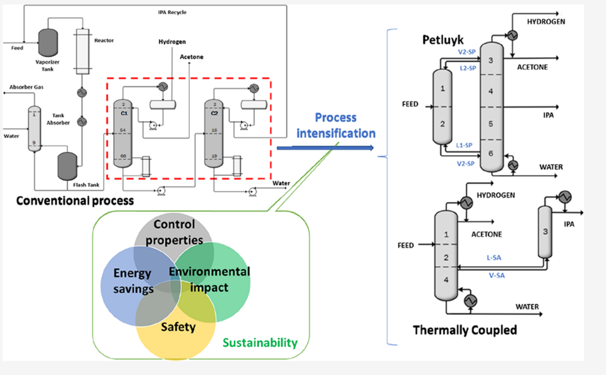
ABSTRACT: Acetone purification is one of the most critical stages of its production process, because a large amount of energy is required. Due to this high energy consumption, the process turns out to be not very sustainable and not friendly to the environment. In this sense, the development of intensified alternatives that minimize energy consumption in this process is of utmost importance. Besides, the safest possible processes are sought, so it is necessary that the control properties of these novel processes be studied at an early design stage. This work proposes two new intensified systems for the purification of acetone; the intensified schemes are a thermally coupled distillation system with a side rectifier and a Petlyuk arrangement. The results indicate that in this type of systems where interconnection flows are used, the magnitudes of these flows have a direct impact on energy consumption, because lower values of interconnection flows as in the case of the thermally coupled system achieve a reduction, whereas higher values as in the case of the Petlyuk system have a negative impact. As for the control properties, the intensified schemes present better values of the condition number with respect to the conventional design, because the interconnection flows reduce the disturbance of the manipulable variables. On the other hand, if the feed is disturbed, the interconnection flows generate an increase in the disturbance in the system, obtaining that the conventional system presents the best values. Therefore, making a balance between the studied designs and looking for a system that presents the best sustainability indicators, the thermally coupled system obtains the best results with a 25.92% energy saving and CO_2 emission reduction with respect to the conventional system and acceptable values for the control and safety indexes.



1. INTRODUCTION

The size of the global acetone market was estimated at USD 4.04 billion in 2018 and is forecast to register a 5.4% compound annual growth rate (CAGR) in terms of revenue during the forecast period. Furthermore, solvent manufacturing consumed 32.9% of the total acetone demand worldwide in 2018.[1] The demand for acetone has increased thanks to end-use industries such as personal care, textiles, electronics, cleaning, pharmaceuticals, and petroleum. Acetone is important in cosmetics because it is one of the most widely used solvents in nail polish removers and cleaners.[2] This use is due to the physical and chemical properties, such as low boiling point and miscibility in water,[3] that drive the consumption of the product and project to boost the world market in the coming years.

Acetone is produced by different routes, such as the oxidation of cumene (Hock Process), the dehydrogenation of 2-propanol (IPA), and the direct oxidation of propylene.[4] It should be noted that 96% of the global acetone production comes as a by-product of phenol production.[5] This route is carried out in two stages. In the first stage of Friedel−Crafts alkylation, cumene is produced with reagents such as benzene and propene. After this, in the second stage, the oxidation of cumene, or Hock process, is carried out, in which two products of great interest (phenol and acetone) are simultaneously synthesized from a single reagent, cumene hydroperoxide.[6] This process is characterized by not adhering to the principles of green chemistry and sustainability because it uses a mineral acid (sulfuric acid).[7] The resulting mixture containing sulfuric acid, phenol, acetone, acetophenone, and sulfonated by-products[8] is neutralized and purified using distillation columns. The operating cost of this separation process is high.

On the other hand, the search for a process that is more friendly to the environment due to the need to incorporate the principles of sustainability as a design objective instead of a

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limitation has caused important changes in the design of chemical processes in the past decades. In this sense, the evaluation of sustainability through economic, environmental, and social dimensions is becoming a means to re-evaluate the design of a certain process or product. However, the inherent complexity of sustainability assessment gives rise to various challenges that must be addressed at each stage of process design. At first, the design problem is open to interpretation, and information is scarce; therefore, the metrics for the sustainability dimensions cannot be easily implemented. In later design stages, when detailed information is available, the potential for sustainability improvement is limited by preliminary decisions made on process design.[9] Therefore, process intensification is a tool that allows the development of new apparatus and techniques that, compared to those commonly used today, are expected to bring drastic improvements in manufacturing and processing, substantially reducing the size/capacity ratio of production, energy consumption, or waste production, and ultimately, produce cheaper and more sustainable technologies.[10]

One of the alternative processes to produce acetone is the dehydrogenation of IPA, whereby an endothermic reaction in the gas phase converts the IPA into acetone and hydrogen.[11] The main advantage of this process is that the acetone produced is free of traces of aromatic compounds, in particular benzene. For this reason, acetone produced from IPA may be preferred in the pharmaceutical industry due to the strong restrictions on using solvents.[12] Likewise, the source of the cumene oxidation process (Hock Process) is oil, while IPA can be synthesized from various sources such as oil, natural gas, or biomass.[13] From the global energy and environmental point of view, acetone production from IPA has CO_2 mitigation benefits.[13] On the other hand, most of the energy to generate hydrogen is generated from coal and natural gas through high-temperature water hydrolysis.[14,15]

Furthermore, to produce hydrogen with zero or low environmental impact ("green" hydrogen), all CO_2 and other pollutants must be processed (i.e., separated) when hydrogen is extracted from fossil fuels.[15] Therefore, the high cost and complicated process of this approach have become an obstacle for mass hydrogen production. Then, the dehydrogenation of IPA is considered a clean and sustainable energy source to produce hydrogen.[16] Therefore, the IPA dehydrogenation process can be seen as an alternative from two perspectives. First, the dehydrogenation of IPA is an alternative route to produce acetone, and second, the dehydrogenation of IPA is a way to produce hydrogen.[16] This process consists of a vaporizer, a tubular reactor, a flash tank, an absorber, and two distillation columns.[11] An important characteristic of this process occurs in the separation stage due to an azeotrope in the IPA/water system.[11] The normal boiling points of IPA and water are 355.20 and 373.17 K, respectively. The azeotrope contains a mole fraction of IPA of 0.647 at 1 atm and 352.72 K. The ternary diagram is available in the Supporting Information.

The processes for the separation of liquid mixtures include many forms of distillation and depend on the vapor−liquid balance of the components in the mixture and their thermodynamics. In the case of purification of non-ideal mixtures (e.g., separation of azeotropes), the separation processes consume an important amount of energy and lead to high capital costs and environmental pollution,[17] which justifies searching for alternatives to conventional distillation, which is normally used for the purification of acetone.[18−20] Likewise, the problems related to the high consumption of energy required by the distillation processes and the need to reduce energy costs have promoted the search for solutions to achieve efficient use of energy in the distillation process.[21−24] Therefore, various authors have reported intensified processes with favorable economic, environmental,[25−28] and energy savings,[29−32] such as thermally coupled systems,[33−36] thermally coupled dividing wall columns,[23,35,37,38] hybrid systems such as reactive distillation,[39−42] and Petlyuk systems,[43−49] among others. The use of intensified distillation processes for multicomponent systems has been recognized as an alternative to reduce energy consumption and operating costs.[48,50] These types of schemes can offer operational advantages compared to conventional distillation systems. However, the modeling and simulation of these intensified processes can be difficult because there are no design methods to effectively determine the optimal values of the degrees of freedom that allow the suitable operation of the separation scheme.[51]

In this study, we extended the acetone recovery process based on conventional distillation to a modified process using thermally coupled configurations. As far as the authors are aware, this type of intensified processes has not been studied, from the point of view of sustainability indicators, for acetone purification. The main contribution of this work is to explore the possibility of using these intensified configurations at an industry level, in a framework of sustainability. These systems provide choices of special interest for the separation of multicomponent mixtures because of their potential energy savings. Also, in this study, the advantages of those intensified configurations are analyzed using energy, control, environmental, and inherent safety indexes as indicators of sustainability.

2. - SUSTAINABLE PROCESS

Technological modernization of chemical plants requires the design of more advanced plants, the development of more efficient information technology, the imposition of more demanding environmental and economic restrictions, and the solution of complex process modeling and control problems. Hence, it is increasingly common to control the highly nonlinear performances of the plants, which require the design of controllers that operate with good performance in a wide range of the operating space[52] with sophisticated control strategies. A few years ago, these advanced strategies were difficult to implement due to computer technology restrictions at that time.[53] Currently, it is important to use indicators for characteristics of the inherent safety of the process to eliminate, reduce, or avoid the sources of risk.[52] The importance of considering sustainability issues early in intensified process design can help differentiate between processes that are easy or difficult to operate. According to Jimenez-Gonzalez et al.,[54] we should consider incorporating "green metrics" when designing an intensified process toward the broader goal of environmental sustainability. Among these green metrics, it is worth highlighting the environmental, health, safety, and process control aspects. Principle 11 in green chemistry expresses the desire for real-time process monitoring and analysis. The goal of this principle is quite simple: prevent waste and safety issues by identifying process excursions as they occur. Thus, it may be high time to modify the process parameters so that the excursion can be reversed and there is no subsequent impact on the safety and quality of

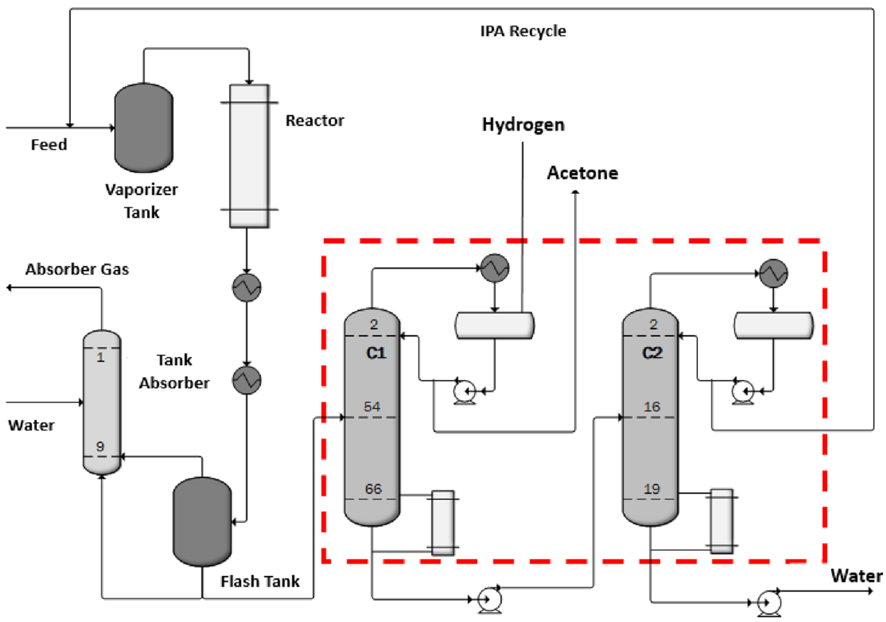

Figure 1. Conventional system. The dotted area represents the part of the process where the intensification will be implemented.

the final product. Real-time analysis and process control are necessary to carry out this action.

In the same sense, the intensification of process processes, associated with the reduction in the number of equipment and the change in the system topology, can also modify the control properties and dynamic performance compared to unmodified systems.[55] This is so that the modifications due to the intensification of the process do not affect the quality of the final product or the safety of the process.[56] This situation is possible only if the process is stable from the point of view of process control, hence the need for a simultaneous study of security and control issues.

Several authors have carried out control studies in systems for obtaining acetone. Kitikiatsophon et al.[57] studied the dynamic behavior of an isopropanol-acetone-hydrogen chemical heat pump system. Luyben et al.[58] studied the control of an azeotropic system for the azeotropic distillation of acetone/chloroform. Luyben et al.[11] developed a control structure of the whole plant capable of effectively handling large disturbances in the production rate. Zhu et al.[59] studied the dynamic control for extractive distillation to purify acetone. Contrary to the conventional distillation process, the dynamics and safety of thermally coupled columns for acetone purification has not been explored in the published literature.

Therefore, in this work, the intensification of processes in the distillation columns was implemented, using thermal coupling, with the aim of reducing the energy consumption of the process. To evaluate the pollution due to the energy consumption of the systems, CO_2 emissions are calculated. To guarantee a safe process, an analysis of the control properties was carried out, using the sensitivity index,[60] to evaluate the behavior before feeding variations, and the condition number,[61] to evaluate the behavior of the systems before the variations of the manipulable variables of the system. In addition, the security metric was calculated through the concept of individual risk (IR) index. The IR can be defined as the risk of injury or decease to a person in the vicinity of a hazard.[62] These criteria were chosen according to the 12 principles of green and sustainable processes proposed by Jiménez-Gonzalez and Constable.[63] Rafiei and Ricardez-Sandoval[64] have also established that the sustainability of processes can be improved by employing a more integral approach where different parameters are considered. This integral approach considers different design variables that affect some parameters of sustainable process such as process controllability, safety, environmental impact, and economy. Hence, a more integral approach where different metrics are considered provides a more extended overview for decision-making procedure to select the most sustainable option process. The calculation of each of the metrics is detailed in the Supporting Information.

3. CASE STUDY

The purification of acetone from the dehydrogenation of IPA with the flows and feed compositions previously defined by Luyben et al.[11] was used, considering a feed flow of 51.96 kmol/h. The flow composition for the feed stream was 67 mol % IPA and 35 mol % water at 320 K and 0.999 atm. This flow was used to have a point of comparison with the design proposed by Luyben et al.[11] All the proposed configurations were simulated using Aspen Plus V11. The properties were estimated with the UNIQUAC thermodynamic model[65,66] for this system that presents several azeotropes. The minimum purity targets were set at 99% for acetone, 98.9% for water, and 62% for IPA in the distillate. Acetone purity was selected based on its use as a solvent.[67−69]

3.1. Synthesis of Intensified Alternatives. The proposed designs were obtained following the methodology shown by Errico.[70] The methodology consists of four steps and assists in the determination of stripping and rectification zones, thermal couplings, intensified sequences, and the location of side streams. In our case, the reduction of one column was not performed; only the first three steps were followed:

- Step 1: Identification of the reference configuration: The first step is to define a reference configuration. In this case, it is the conventional distillation system.
- Step 2: Generation of modified thermally coupled configurations: The crucial part of the present stage is

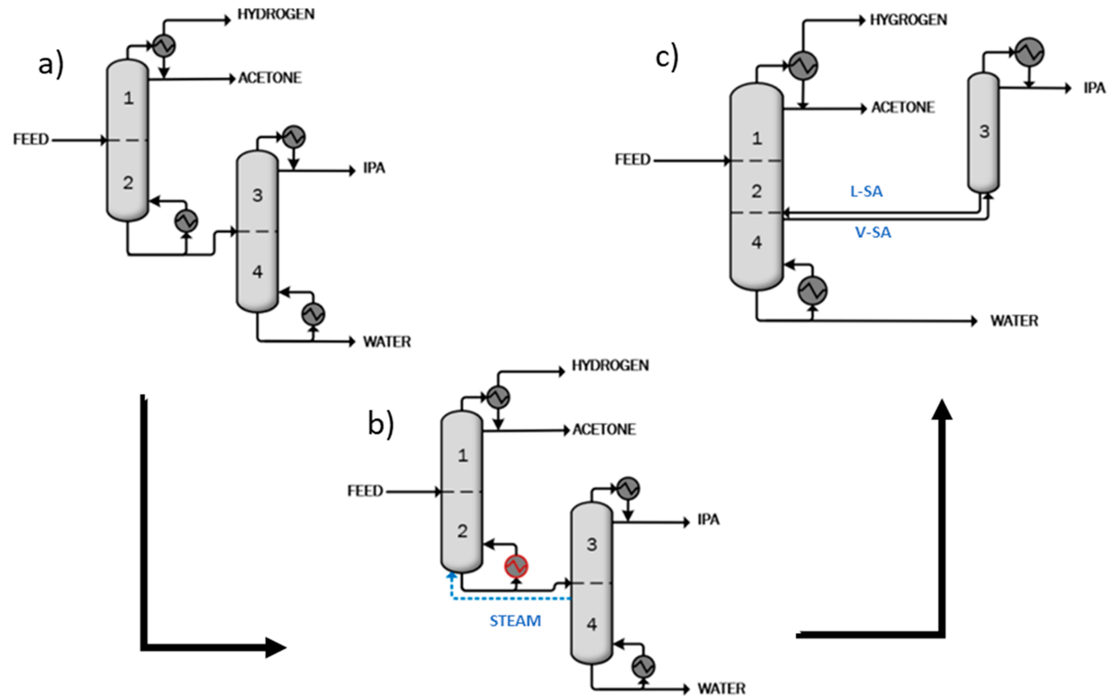

Figure 2. Thermally coupled distillation system.

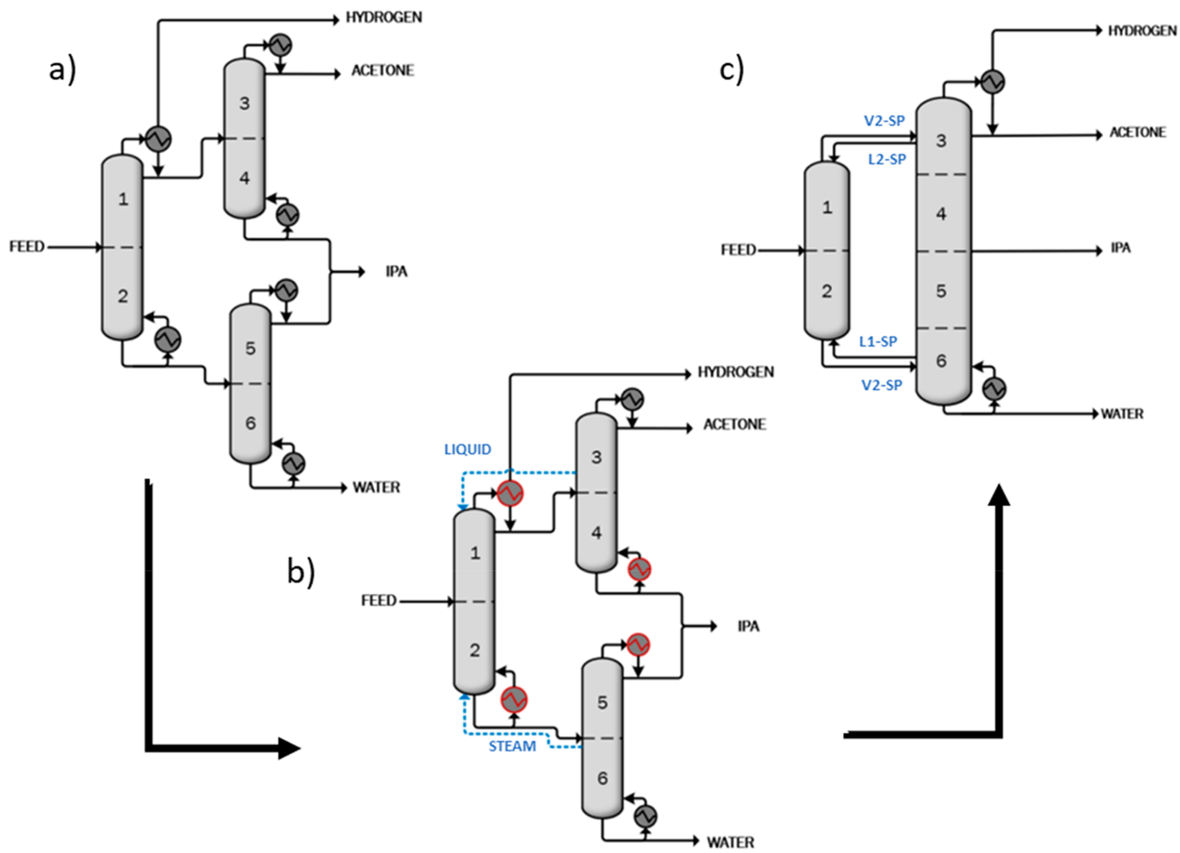

Figure 3. Petlyuk system.

to generate the modification of the reference configuration by replacing the auxiliary equipment (reboiler/condenser) with interconnection streams (liquid–vapor). Such equipment is replaced by interconnection streams, leaving room for multiple options.

- Step 3: Identification of thermodynamically equivalent configurations: It is important to consider ideal mixtures as this will help generate thermodynamically equivalent configurations with the movement of column sections. However, because it is an azeotropic mixture, it is not so

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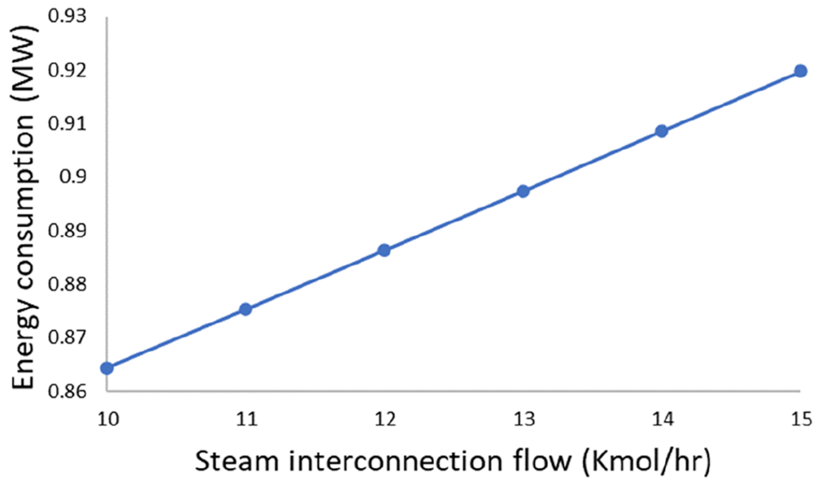

Figure 4. Effect of the interconnection flow in the reboiler duty for the thermally coupled system.

simple. Several possible configurations can be obtained by moving the individual sections simultaneously or independently.

In this specific case, the initial stage of the synthesis of this intensified process was carried out by sequentially applying thermal couplings. The intensified distillation schemes proposed are a thermally coupled system and the use of a Petlyuk system. The schemes studied are shown below:

a Conventional distillation system: This design is considered to have a point of comparison with the intensified schemes; this is based on the reported by Luyben et al.,[11] in which 2 distillation columns (C1 and C2) are considered (Figure 1).

b Thermally coupled distillation system: Starting from the columns of the conventional system (Figure 2a), thermal coupling is performed on the part of the bottoms (blue dotted line; Figure 2b). Subsequently, a movement of sections is carried out, resulting in our thermally coupled system (Figure 2c). The L-SA and V-SA flows correspond to the liquid and vapor interconnection flows, respectively.

c Petlyuk system: Starting from the columns of the conventional system, a flow redistribution was performed, and a column was added to the system, as shown in Figure 3a. A pair of thermal couplings were made between the first column and the two subsequent ones; both couplings are represented by the blue dotted lines in Figure 3b, generating interconnection flows of liquid (L1-SP and L2-SP) and interconnection flows of steam (V1-SP and V2-SP). Subsequently, the movement of sections is carried out, which was carried out by joining Section 4 (rectification zone) and Section 5 (stripping zone), combining the stages of both columns, resulting in the Petlyuk system, as shown in Figure 3c. For a better association between the designs, for the three schemes, the first column was named C1 and the second column C2.

Starting from the intensified schemes, the sensitivity analysis was carried out for the proposed systems, with the aim of finding the values of the process variables that would minimize the reboiler duty because thermally coupled systems do not provide an initial value for the currents of interconnected liquid or vapor. Therefore, these currents must be specified and optimized because interconnection flows have been determined to be one of the most important design variables because their values have direct effects on energy needs. Therefore, non-optimal values of these parameters can lead to low energy savings for thermally coupled systems or even higher energy consumption compared to their conventional counterparts.[71−73] Therefore, in the case of the conventional system, the values of the design parameters reported by Luyben et al.[11] For all designs, the same purity restrictions of the products (acetone, IPA, and water) were considered. In this sense, initial values for interconnection flows can be obtained from conventional schemes.

To carry out the optimization of the interconnection flows for the thermally coupled and Petlyuk systems, it must be considered that acetone, water, and IPA must be recovered with a molar purity of 99, 98.9, and 62% respectively. It is important to note that the optimization of interconnection flows must be done considering the same product compositions in order to achieve a representative comparison of the energy requirements for different interconnection flows. The variations in interconnection flows were made in a staggered manner, with an increase of 0.001% with respect to the nominal value, due to the fact that the system is highly nonlinear. Once the sensitivity analysis results in the reboiler duty being obtained, the emissions generated due to the energy required to generate the heating services were calculated.

Starting from the optimal models of each studied system, a control study was carried out at zero frequency. For this, a disturbance was implemented starting from the nominal operating point, with a magnitude of +0.5% in a stepwise manner to +5% of each selected manipulative variable. The size of this disturbance was obtained through a sensitivity analysis of the system, taking as a criterion that, with a minor disturbance, the influence of nonlinearities is avoided.[74] Similar studies have been presented in a dynamic state that have shown that intensified systems are sensitive to small disturbances.[75−77] Last, the quantitative risk analysis (QRA) methodology is used to calculate the IR index. The calculation of each of the metrics is detailed in the Supporting Information.

4. RESULTS

This section presents the results of the sensitivity analysis to minimize the reboiler duty of the intensified systems, as well as

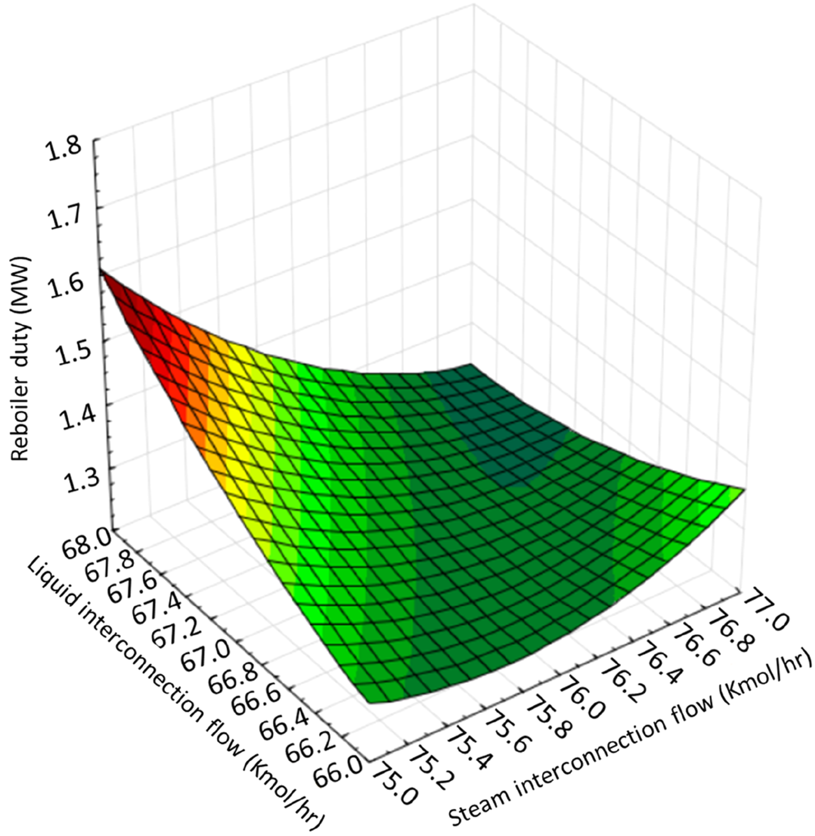

Figure 5. Effect of the interconnection flow in the reboiler duty for the Petlyuk system.

an analysis of the control and safety properties using as indicators the condition number and the sensitivity index and Induvial Risk in the three systems studied.

4.1. Results of Sensitivity Analysis to Minimize Reboiler Duty. For this analysis, a sensitivity analysis of the interconnection flows was carried out in the proposed intensified designs, taking as a criterion the minimization of the reboiler duty. Design parameters such as the number of stages, feed stage, diameter, output flows, and required purities were kept constant. For the thermally coupled system, the steam interconnection flow (V-SA) that is fed from C1 to C2 was manipulated. For the Petlyuk system, both the L2-SP liquid interconnection flow as well as the V1-SP vapor interconnection flow were manipulated. For both the thermally coupled system and the Petlyuk system, the reflux ratio was manipulated in such a way that in each variation of the interconnection flows the concentrations and recoveries in the flow rates were maintained.

Figure 4 shows the results of the thermally coupled system in terms of the variation of steam interconnection flow and its impact on the reboiler duty, so this system presents a linear behavior to the variations of V-SA. The minimum value reached in the reboiler duty was 0.86 MW with an interconnection flow of steam of 10 kmol/h. At lower flows of V-SA, the system becomes unstable due to redistribution of flows and thermodynamic equilibrium. In the results of the variation of the L2-SP and V1-SP flows, in the Petlyuk system, the variation intervals were set based on a sensitivity analysis, where the system did not lose convergence and where the reboiler duty showed the lowest values. The value of the interconnection flows of steam and liquid were 76.7 and 67.9 kmol/h, respectively, with the minimum reboiler duty reached at a value of 1.26 MW, as shown in Figure 5.

The design parameters of the resulting schemes are shown in Table 1.

Comparing the conventional design with the thermally coupled design, we can see that in column C1, the reflux ratio is lower for the thermally coupled system and, thus, the reboiler duty is favored. In the case of column C2, the number of stages is reduced, and with this, the reflux ratio is increased. Based on a global balance of both designs in terms of energy consumption, the thermally coupled system presents an energy saving of 25.92% compared to the conventional system. Using thermal coupling, a remixing effect is promoted, and by taking advantage of the temperature of the V-SA flow for the separation of the IPA, it is possible to reduce energy consumption.

In the Petlyuk system, C1 is smaller compared to the other designs, but C2 is a larger unit in terms of the number of stages and diameter, the diameter being the one that has a negative effect on the distribution of the internal flows, increasing the re-flow ratio and generating large interconnection flows. Consequently, there is an increase in energy requirements of 8.64% compared to the conventional system.

It can be concluded that, in this case study, regarding energy consumption, the values of the interconnection flows play an important role in the impact on the reboiler duty. If these values are small, they can favor energy savings due to the remixing effect, such as the case of the thermally coupled system. Nevertheless, if these interconnection flows are large, they negatively affect energy consumption due to increased internal flows, as in the Petlyuk dystem. Figure 6 shows a comparison of the energy requirements of the schemes studied.

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Table 1. Design Specifications for Distillation Sequences

	parameters	systems: conventional	coupled	Petlyuk
C1	number of stages	66	71	17
	feed flow (kmol/h)	72.92	73.05	72.98
	feeding stage	54	54	10
	reflux ratio	2.78	1.62	
	bottoms rate (kmol/h)	40.61	34.78	
	reboiler duty (MW)	1.06	0.86	
	V-SA (kmol/h)		10	
	output stage V-SA		67	
	feeding stage L-SA		66	
	diameter (m)	0.74	0.63	0.40
C2	number of stages	19	16	35
	feed flow (kmol/h)	40.61		
	feeding stage	16		
	reflux ratio	0.45	0.70	2.61
	bottoms rate (kmol/h)	34.85		34.82
	reboiler duty (MW)	0.09		1.26
	feeding stage V-SA		16	
	L2-SP (kmol/h)			67.9
	output stage L2-SP			11
	V1-SP (kmol/h)			76.7
	output stage V1-SP			32
	feeding stage V2-SP			12
	diameter (m)	0.19	0.23	0.69
	total reboiler duty (MW)	1.17	0.86	1.26
	% energy saving		25.92	−8.64
	CO_2 production (kg/h)	211.46	156.67	229.72

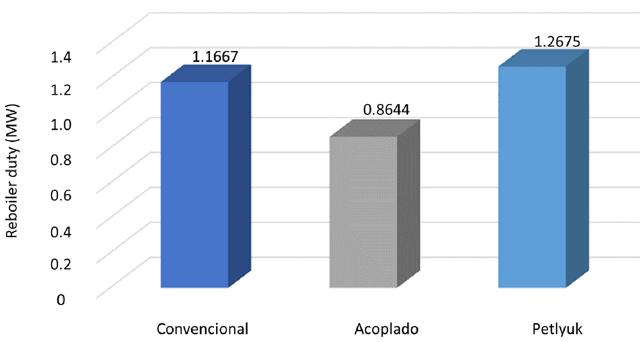

Figure 6. System reboiler duty.

These results agree with those reported by Tedder and Rudd,[79] where the best alternatives for the separation of ternary mixtures are presented. In the work of Tedder and Rudd, the best alternatives are selected according to the composition of the feed, thereby guaranteeing the best design in terms of energy savings. Within two of these rules, the proposed designs are mentioned:

- If 40 to 80% is the middle product and nearly equal amounts of overhead and bottoms are present, then please design Petlyuk.
- If less than 15% is the middle product and nearly equal amounts of overheads and bottoms are present, then please design direct thermally coupled.

In this case, because the amount of IPA (middle product) is less than 15% and almost equal amounts of overheads (acetone) and bottoms (water) are present, the configuration that favors separation is direct thermally coupled. Therefore, this means that to separate this mixture with this specific composition, the Petlyuk system does not represent the best option from its design stage. Although this can be seen from the design stage, it is important to point out that these rules are heuristics, so it is necessary to analyze the proposed designs and their feasibility, especially knowing the energy advantages that the use of the Petlyuk system can bring and the possibility of presenting better control and safety properties.

4.2. Results of CO_2 Emissions. Table 1 provides the results of the calculation of the CO_2 emissions produced to satisfy the energy supplied to the reboilers of each process. The savings of the coupled system with respect to the use of natural gas is 27% compared to the conventional system. The Petlyuk system, presenting a higher energy consumption, has an increase of 9% compared to the conventional system. As expected, the coupled system has an emission saving of 27% compared to the conventional system; this represents more than 370 tons per year, if we take into account that the average CO_2 emission per vehicle in the world is 4.6 tons per year;[78] this reduction in CO_2 emissions would be equivalent to eliminating pollution from 80.5 vehicles in 1 year.

4.3. Results of Control Properties. To carry out the control analysis of the systems, the manipulable variables that directly affect an output variable were identified, the output variables being the purities of acetone, IPA, and water. Therefore, for the condition number (Table 2), the manipulable variables associated with the purities of the products of interest were disturbed.

Table 2. Condition Number Results

percentage of disturbance (%)	systems: conventional	coupled	Petlyuk
0.5	3.06×10^3	1.47×10^2	3.11×10^1
1.0	3.08×10^3	7.70×10^1	2.10×10^1
1.5	3.09×10^3	7.25×10^1	2.30×10^2
2.0	3.40×10^3	4.66×10^1	5.50×10^2
2.5	5.30×10^3	5.87×10^1	1.37×10^2
3.0	6.83×10^3	8.19×10^1	8.48×10^0
3.5	7.80×10^3	9.86×10^1	6.54×10^0
4.0	7.61×10^3	1.18×10^1	3.96×10^0
4.5	8.16×10^3	1.34×10^2	3.06×10^0
5.0	8.57×10^3	1.43×10^2	2.70×10^0

For the conventional system, the control arrangement for C1 was the reflux ratio because this current is directly associated with the purity of the acetone that comes out through the dome; for C2, the flow of the bottom stream associated with the purity of the water and having a partial condenser, the reflux ratio was chosen as the variable associated with the purity of IPA. In the thermally coupled system in C1, the reflux ratio is associated with the acetone composition and the flow of the bottom stream to the purity of the water. For C2, the reflux ratio is associated with the IPA purity. Finally, for the Petlyuk system, manipulable variables for C2 are considered the reflux ratio associated with acetone purity, bottom flow associated with water purity, and intermediate IPA flow associated with IPA composition.

On the other hand, for the sensitivity index, the behavior of the system is evaluated, considering the same output variables, but the manipulable variables of the feed, which were the feed flow and the IPA composition, were disturbed. The objective of implementing the condition number and the sensitivity

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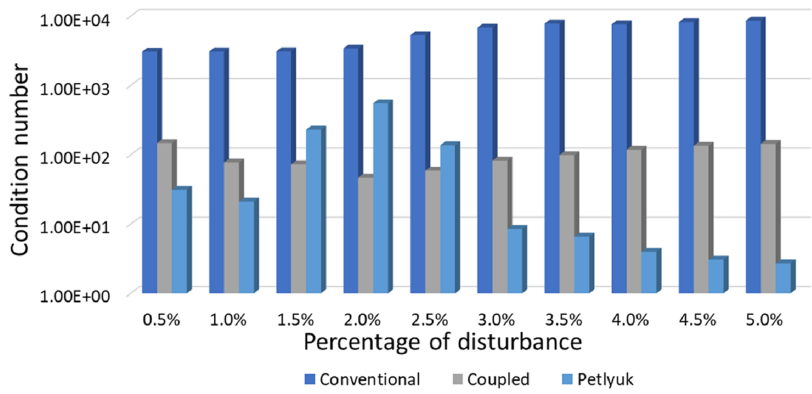

Figure 7. Condition number results.

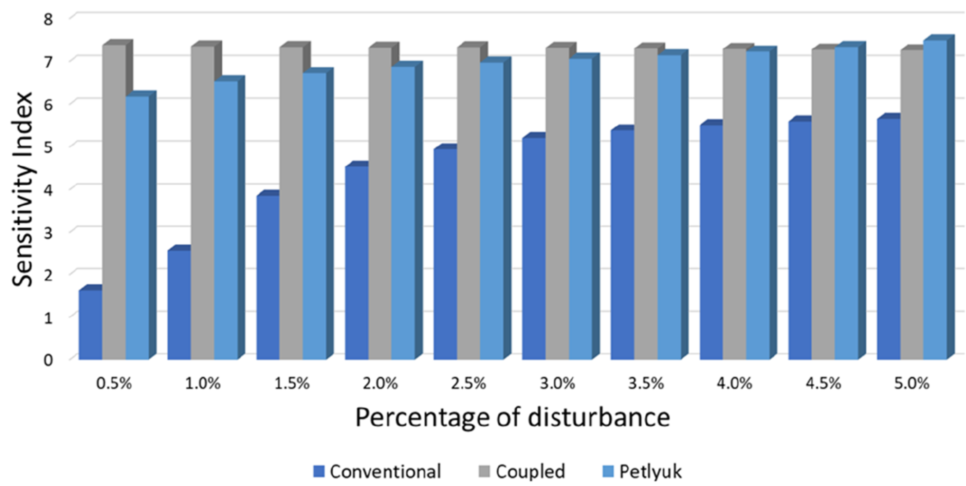

Figure 8. Sensitivity index results.

index is to evaluate how sensitive the output variables are to disturbances to the system.

The results of the condition number are shown in Figure 7. The thermally coupled system does not present the best condition number values among the disturbance values bounded between 1.5 and 2.5% (Table 2). The Petlyuk system presents the best values in most disturbances. However, the thermally coupled system has lower values than the conventional system. The conventional system is more unstable and presents the largest disturbance values because it does not have recycles that allow dampening disturbances. Finally, the Petlyuk system presents the best values at large disturbances because the recycles present in its structure help the system eliminate disturbances. In this case, the value of the interconnection flow rates minimizes the impact of disturbances better than the thermally coupled system, which has lower interconnection flow values than the Petlyuk system.

Regarding the results for the sensitivity index, as shown in Figure 8, the minimum values are those associated with the conventional system because in this system the disturbance in the power supply propagates linearly from C1 to C2. On the other hand, the disturbance in feed flow rate enters column C1 in all systems, and in the case of the conventional system C1, it has the largest characteristic diameter that allows it to dissipate the disturbance better. If we compare the thermally coupled system with the Petlyuk system (Table 3), it is observed that at disturbances lower than 4%, the coupled system is more sensitive to disturbances at the input; at disturbances higher than 4%, the system has a lower impact on the coupled system. Because the Petlyuk system has a double recycling, the effect is reduced at low disturbances, but at higher disturbance, this double recycling generates a snowball effect that increases the sensitivity. On the other hand, recycling in the thermally coupled system has an inverse effect than in the Petlyuk system, generating a lower sensitivity to greater disturbances.

Table 3. Sensitivity Index Results

	systems		
percentage of disturbance (%)	conventional	coupled	Petlyuk
0.5	1.63	7.38	6.18
1.0	2.56	7.35	6.54
1.5	3.86	7.33	6.72
2.0	4.54	7.32	6.88
2.5	4.65	7.33	6.97
3.0	5.21	7.32	7.06
3.5	5.39	7.30	7.14
4.0	5.51	7.29	7.23
4.5	5.60	7.27	7.34
5.0	5.65	7.26	7.49

The intensified schemes have better control properties. If we consider the results of the condition number, the Petlyuk system is the one with the best values, followed by the thermally coupled and finally the conventional system. The conventional design presents the best values regarding the

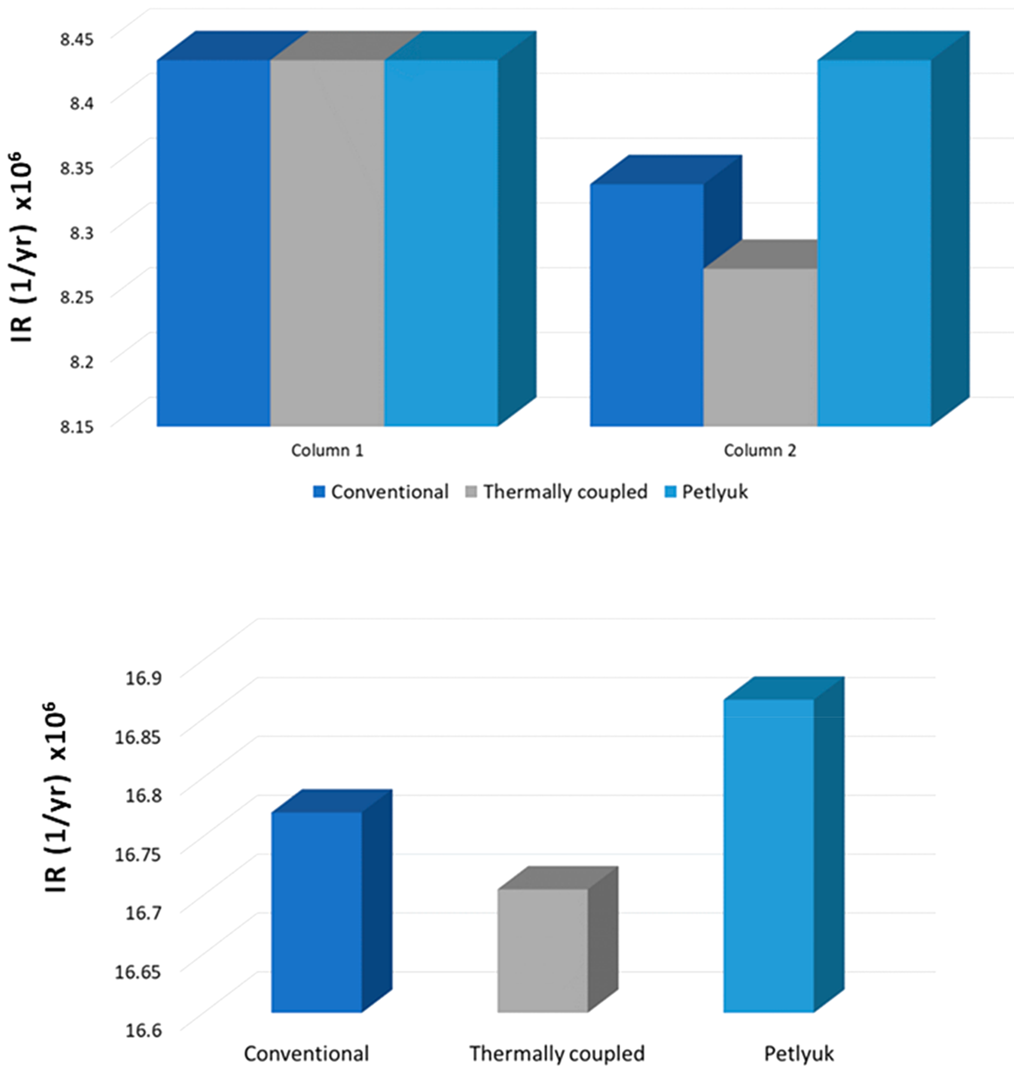

Figure 9. Safety index results.

Table 4. Summary of Indexes Results

	reboiler duty (MW)	saving of: energy, CO_2 emissions and natural gas	condition number (5% percentage of disturbance)	IS (5% percentage of disturbance)	(1/yr) × 10^6
conventional	1.16		8.57×10^3	5.65	16.77
coupled	0.86	25.92	1.43×10^2	7.26	16.70
Petlyuk	1.02	−8.64	2.70×10^0	7.49	16.86

sensitivity index, and the intensified schemes present similar values.

4.4. Results of Process Safety. The results of safety analysis of all columns are shown in Figure 9. It is important to highlight that this process contains mainly organic compounds and hydrogen; these chemicals are volatile and flammable; for those reasons the inherent safety of the process is an important metric to be considered. In this work, only the safety properties of distillation columns are evaluated because the topology and concentrations of other equipment were kept constant. This was done with the aim of only evaluating the effect of distillation columns and their topology on the safety of the process. Please note that the prefractionation column of Petlyuk configuration is named column 1, whereas the rotifer of thermally coupled sequences is called column 2 in order to simplify the comparison.

Based on the results of Figure 9, it is evident that the intensification does not have any impact on the safety of the first column. Although this column has different number of stages, diameters, and heights, the high concentration of organic compounds and hydrogen in the feed stream has the most important effect on the safety index. The effect of the concentrations of organic compounds and hydrogen is so pronounced that the effect of the column size for the different intensified processes on safety index is practically negligible. However, the topology of distillation processes plays a key role in the safety index for the second column. In this case, the Petlyuk configuration has the worst safety performance because this configuration separates all the compounds in the second column, which means that the high concentrations of organics and hydrogen are also present in the second column, which worsens the safety. Therefore, it is concluded

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that the Petlyuk arrangements are not the best option to purify mixtures rich in organic compounds from a safety point of view. On the other hand, the thermally coupled system showed the best safety properties in contrast to the conventional and Petlyuk arrangements. The configuration is the safest because its second column (rectifier) has a low concentration of organic compounds, less number of stages, and therefore smaller size, which causes the consequences of an accident in this column to be less severe. Finally, the conventional column has better safety than the Petlyuk configuration but worse than thermally coupled. This configuration separates acetone and hydrogen in the first column, which notably decreases the concentration of organic compounds in the second column.

Table 4 provides the summary of the results of the indices obtained. In terms of energy saving, the thermally coupled system presents a saving of 25.92% compared to the conventional scheme, which has an equivalent impact on its natural gas consumption and CO_2 emission. Moreover, balancing the studied designs and looking for the system that presents the best sustainability indicators, the thermal coupling system obtains appropriate values of the control indices and the lowest value of the safety index.

The results obtained during this analysis resemble the guidelines reported by Jimenez-Gonzalez et al.,[54] where it is mentioned that the process that presents the best energy savings will have the best control properties and will be the safest. Accordingly, the thermally coupled system represents the most sustainable design of the studied systems. In this way, a sustainable intensified process is generated to produce acetone.

5. CONCLUSIONS

In this work, different alternatives were designed to purify acetone and by-products, such as hydrogen, starting from the production process through the dehydrogenation of IPA. The intensified processes were contrasted with their respective conventional option, and the results indicate that a significant reduction in the reboiler duty of the separation process of 25.92% can be achieved through the thermally coupled system, which is associated with a 25.92% reduction in CO_2 emissions. Likewise, this system contains stable control properties before disturbances. Although this system does not present the best control characteristics, its behavior in the face of different disturbances is stable. On the other hand, significant reductions in operating costs and improvements in separation are achieved given the previously mentioned energy and control characteristics. It is extremely important to highlight this due to the azeotrope present between the IPA and the water. Given this, the factor with the greatest contribution and impact to consider is the reboiler duty. Thus, in this case, although the coupled system is not the best design in terms of controllability, its energy consumption presents relevant savings. On the other hand, this system has a smaller diameter than the other two systems, so although this design would present an economic saving in operation; it would also present it in the investment. Therefore, in terms of a balance between the studied designs and a system that presents the best sustainability indicators, the system with thermal coupling obtains the best results with a reduction of 25.92% in energy consumption compared to the conventional system with acceptable values in the control indexes. Thus, the thermal coupling has better safety indicators from a control point of view than the conventional system, and it is a sustainable process to produce acetone. The results of safety index indicate that the intensified processes have different safety properties than conventional process. However, these safety properties are not necessarily better than the conventional process. Therefore, whether an intensified process is more or less safe will depend on the distribution of concentrations of flammable and toxic compounds within the equipment, which is directly related with the topology of the intensified process.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.iecr.1c04321.

Ternary diagram, calculation of the CO_2 emissions, procedure for calculation of condition number, procedure for calculation of sensitivity index, and procedure for calculation of risk index (PDF)

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Notes

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■ ABBREVIATIONS

CAGR compound annual growth rate
IPA 2-propanol
GHG greenhouse gas
L-SA liquid interconnection flow in thermally coupled system
V-SA vapor interconnection flow in thermally coupled system
L1-SP first liquid interconnection flow in Petlyuk system
L2-SP second liquid interconnection flow in Petlyuk system

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V1-SP	first vapor interconnection flow in Petlyuk system
V2-SP	second vapor interconnection flow in Petlyuk system
γ	condition number
SI	sensitivity index
HHV	higher heating value
IR	individual risk
QRA	quantitative risk analysis
HAZOP	hazard and operability study
LC50	lethal median concentration

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