



## EVALUATION OF THE ECO-EFFICIENCY OF PROPYLENE AND PROPANE DISTILLATION PROCESSES

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**Abstract:** Propylene is used in the manufacture of various materials such as polypropylene, acrylonitrile, cumene, and acrylic acid. Its production is promoted by cracking, which generates light hydrocarbons as by-products, requiring further purification. Of all purification processes, the distillation of propane-propylene is considered one of the most energy-intensive. Thus, to improve the eco-efficiency of this separation, two intensification strategies by vapor recompression were proposed for two conventional processes of propane-propylene distillation, one with two columns of 100 stages called P1 and the other with a single column of 150 stages called P2, both presented in the literature. To evaluate these processes, eco-efficiency parameters represented by the indicators of water consumption, CO<sub>2</sub> emission, and specific energy cost were quantified, based on calculations made from the results of computer simulation and economic analysis, including the utility plant. These indicators were evaluated by The Eco-efficiency Comparison Index methodology. Results showed that the process P1 is 27,82% more eco-efficient than the process P2. The proposed intensification of P1 process is 28,56% more eco-efficient than the intensification of the process P2, being, in general, 92,70% more eco-efficient for propylene-propene separation.

**Keywords:** Propene, Process intensification, Eco-efficiency, Computer simulation, UniSim.

### 1 Introduction

Propylene is an essential chemical product from the petrochemical industry due to its application in polypropylene, cumene, acrylonitrile, and acrylic acid manufacture. Over the decades, there was an increase in the world demand for propylene, which became one of the most crucial petrochemical products with an estimated production capacity of 114 million tons in 2015 and a projected annual growth rate of 5% (COOMBS, 2016; HORNCastle *et al.*, 2014). Approximately 85% of the world's demand for propylene is provided by thermal cracking and fluid catalytic cracking (CHRISTOPHER *et al.*, 2017). These methods consist of decomposing large hydrocarbon chains into smaller chains thermally or with the help of catalysts (GREENSFELDER *et al.*, 1949). However, these processes have low selectivity, which produces by-products, such as the mixture of propane and propylene that must be further purified.

A variety of methods can be applied to separate chemicals, including extraction (SAMBROOK; RUSSELL, 2006), recrystallization, membrane systems (FREEMAN *et al.*, 2006), and distillation. Thought their low thermodynamic efficiency, distillation is the most common technology for separating chemicals (KAZEMI *et al.*, 2018), and it is typically employed in the purification of propylene and propane.

The distillation unit operation alone is responsible for 10-15% of the total energy consumption in the world globally (SHOLL; LIVELY, 2016). Moreover, according to the U.S. Department of Energy, the separation of propylene and propane is one of the commercial distillations with the highest energy demand (JARVELIN; FAIR, 1993). There are two categories of distillation for this purification: high pressures and lower pressure. High-pressure distillation is carried out at pressures around 1500 kPa to facilitate separating the mixture of propane and propylene (ALCÁNTARA-AVILA *et al.*, 2014). In the condenser, water is used for cooling fluid, and in the reboiler, as the bottom temperature reaches 70°C, it is possible to use low-pressure steam (LIAO *et al.*, 2001). The most significant disadvantage of this process is the high reflux ratio and the high number of theoretical plates.

Furthermore, increasing the operating pressure of the column can considerably raise the energy demand of the boiler (MANN *et al.*, 1963). Low-pressure distillation has operational pressures lower than 1200 kPa, providing high relative propane and propylene volatility, reducing the number of theoretical plates and the reflux ratio (ALCÁNTARA-AVILA *et al.*, 2014). On the other hand, this distillation requires cryogenic temperatures in the condenser, which entails a monetary disadvantage than the high pressure process, making it less applied in the industry (CHRISTOPHER *et al.*, 2017).

Due to the energy-intensive nature of the propylene-propane separation, numerous technologies were developed to substitute the conventional distillation, such as membrane separation (PAN *et al.*, 2012), extractive distillation (LIAO *et al.*, 2001), and adsorption (KHALIGHI *et al.*, 2014). Although the increase in new technologies for this separation in the literature, distillation remains the primary separation process for propylene and propane (CHRISTOPHER *et al.*, 2017). Therefore, enhancement must be done in the process's energy efficiency to align it with the new environment measures made by the World Business Council for Sustainable Development (WBCSD) (MADDEN *et al.*, 2005).

On this basis, the work proposed applying a process intensification of vapor recompression column (VRC) to the distillation of propane and propylene, resulting in a significant reduction of energy consumption and operation cost of the process.

## 2 Process Description

### 2.1 Conventional Processes

The conventional process 1 (P1) consists of 3 steps: the feed (FEED), the storage process, and the distillation, as illustrated in Fig 1. In the distillation are used two 100-plate columns since, as stated by the reference authors (SEIDER *et al.*, 2016), a single 200-plate column would be too large.

The process feed consists of a steam stream. This mixture goes to a storage process, in which compression is carried out by stages, using condensers to control the temperature of the fluid. In the storage tank, there is a vent stream used as a relief for unusual situations.

After storage, the mixture is pressurized in pump B-1 at 2068 kPa, and it is sent to the distillation process. The first column (T-1) receives the fluid in its 62<sup>nd</sup> stage, and it has bottom and top pressures of 2068.43 kPa and 1999.48 kPa, respectively. The liquid from the first stage of the T-1 column is routed to the column system reboiler, while the top steam is directed to the first stage of column T-2. The T-2 column operates with a bottom pressure of 1999.48 kPa and a top pressure of 1930.53 kPa. The bottom product from the T-2 column is redirected to the T-1 column using the B-2 pump, while the top stream is directed to a total reflux condenser, which operates at a ratio of reflux of 15.9.

The conventional process 2 (P2) is the simplest among the studied alternatives, having only one distillation column in the configuration proposed by Seader *et al.* (1998). In this work, to have a fairer comparison with the P1 configuration, a compression stage and storage system similar to P1 was added to the P2 process. It was adjusted according to the operating conditions presented by Seader *et al.* (1998). The flowchart adapted from the P2 configuration is described in Fig 2.

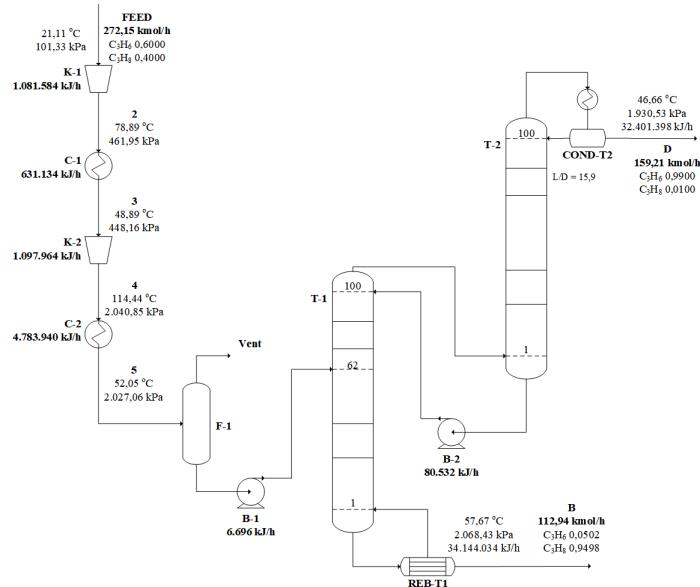


Figure 1: Flowchart of process P1 for the separation of propylene-propane.

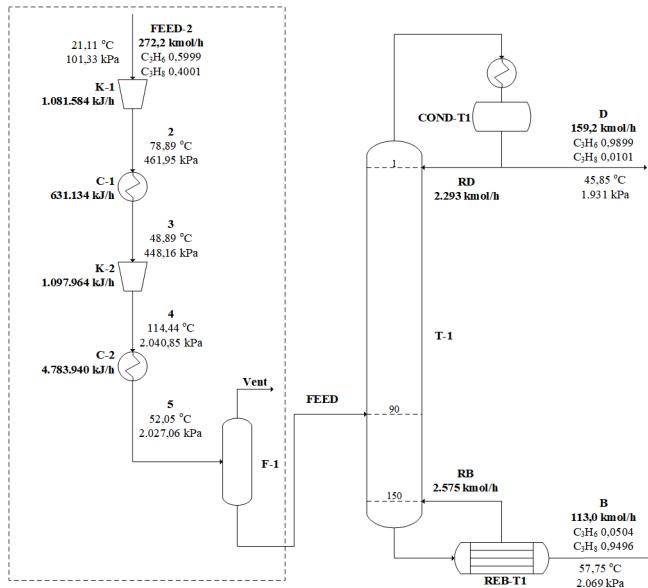


Figure 2: Flowchart of process P2 for the separation of propylene-propane.

The supply stream from Seader *et al.* (1998) work was introduced into the storage system under the same operating conditions as the P1 process feed. Since the specifications proposed in Seader *et al.* (1998) have already been obtained, there was no need to include a pump at the column inlet.

The FEED is introduced into the T-1 90<sup>th</sup> plate, which has 150 plates and operates at pressures of 2069 kPa and 1931 kPa at the bottom and top, respectively. The COND-T1 heat exchanger is a partial condenser with a reflux of 2293 kmol/h. At the bottom of the column, a boiler heats the fluid. The output streams from the T-1 tower are removed in the saturated liquid state.

## 2.2 Intensification Proposal

The proposals to enhance P1 and P2 processes thought vapour represssation strategy were named, they were called VRP1 and VRP2, respectively. They are represented by Figs 3 and 4.

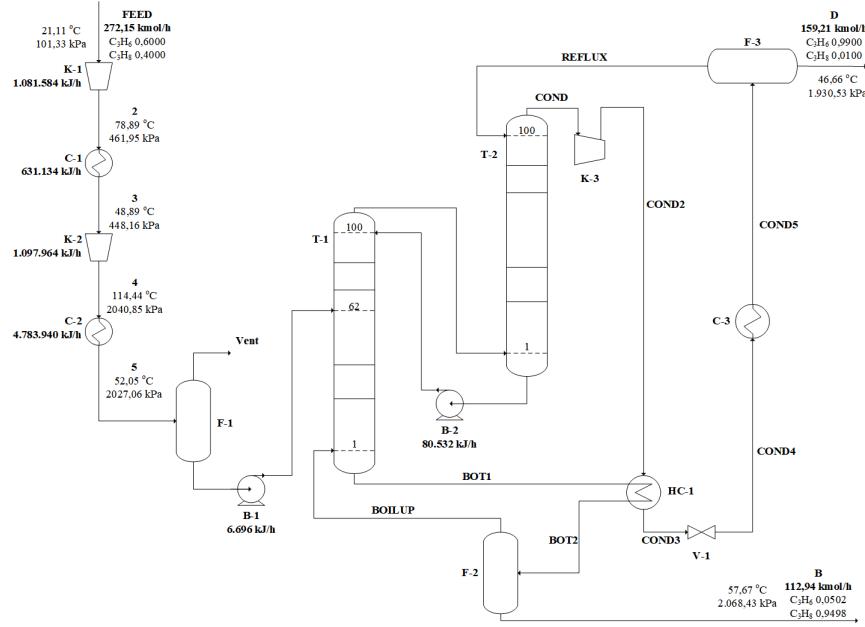


Figure 3: Flowchart of the VRP1 intensification proposal.

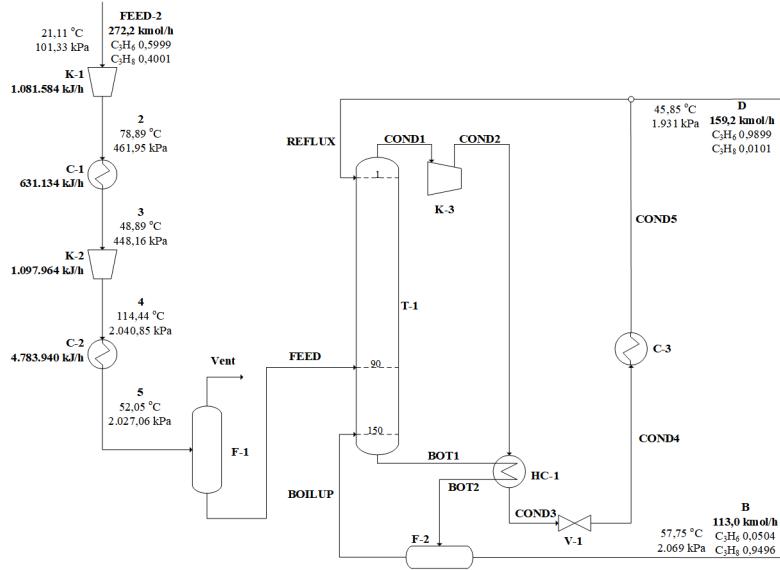


Figure 4: Flowchart of the VRP2 intensification proposal.

The vapor recompression intensification method was used in the reference processes according to each technology's operating conditions and equipment configurations. Adjustments made at the two plants include replacing the reboilers with shell and tube heat exchangers (to allow for heat exchange between the bottom and top products of the columns) and the addition of a compressor for heating the overhead streams. Furthermore, in both proposals, the temperature for the bottom product is reached

without heat exchange utilities.

In addition, the loss of head in the new shell and tube exchangers was neglected, also stipulating a minimum approach of 10°C between the streams, according to the process design heuristics (SEIDER *et al.*, 2016), preventing the exchangers from being designed with high heat exchange areas, which would increase the acquisition cost.

### 2.3 Utilities Plant

In the distillation processes of propylene and propane, water is used as a heating and cooling fluid in the columns reboilers and condensers, presented in the Fig 5. The steam generation system simulation is intended to determine the water consumption and CO<sub>2</sub> emissions of each process. For that, some heuristics were stipulated that describe its specifications according to its pressure as shown in Table 1.

Table 1: Cooling water system heuristics.

Properties	Heuristics value	Reference
Tower inlet temperature	40°C	Turton <i>et al.</i> (2016)
Tower outlet temperature	30°C	Turton <i>et al.</i> (2016)
Heat exchangers inlet pressure	500 kPa	Turton <i>et al.</i> (2016)
Blowdown	3% of the system flow	Turton <i>et al.</i> (2016)
Evaporation lost/Drift	1% of the system flow	Walas <i>et al.</i> (2012)
Lost in the process	1% of the system flow	Seider <i>et al.</i> (2016)

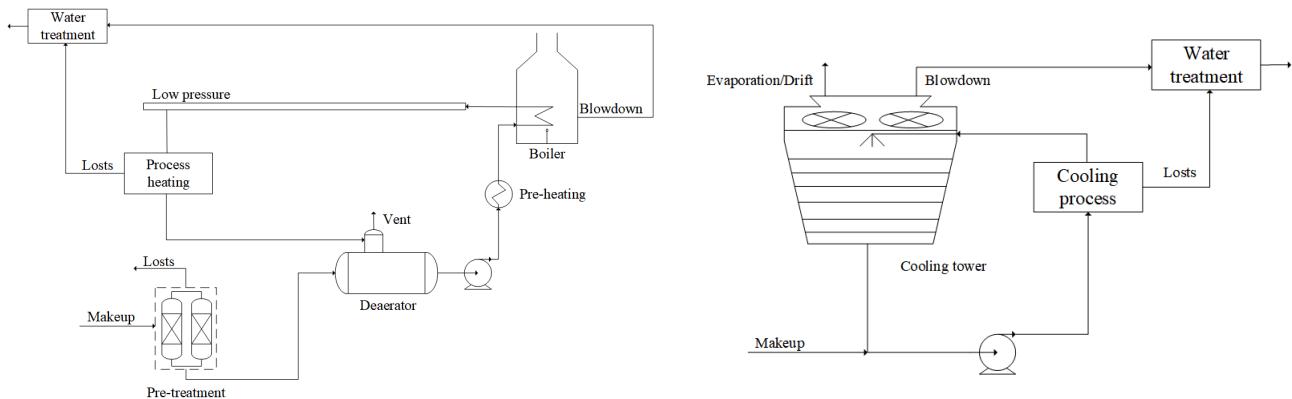


Figure 5: Flowchart of the steam generation system and the open cooling water system with recirculation.

The under stream of the studied separation processes reach a maximum temperature value of 60°C. This allows for the steam generation system operate at low pressure steam (451.3 kPa and 148°C) to be used as a heating fluid. Again the recommendations of 10°C minimum temperature difference between the currents in the heat exchangers was apply (SEIDER *et al.*, 2016).

In the cooling water system, the heuristic of Turton *et al.* (2016) for the system tower inlet temperature was considered in a range of 40-45°C. As the condensers of the distillation columns of the processes operates at a temperature of 46.66°C, the temperature of 40°C had to be considered, aiming the closest possible approximation to the heuristic of a minimum difference of 10°C between the currents in the heat exchangers.

### 3 Methodology

#### 3.1 Process Simulation

The propylene-propane distillation was simulated using the *software* UniSim® Design Suite of *Honeywell* in a steady state condition. Since propylene and propane are the only substances present in the processes studied in this work, the Soave–Redlich–Kwong equation (SRK) was chosen as the thermodynamic model. This package can be applied to a range of non-polar compounds. In the literature there are examples of the application of the SRK package in other intensification methods for the separation of propene and propane, as observed in Mauhar *et al.* (2004) and Ho *et al.* (2009), which corroborates to the thermodynamic model selection for the simulations.

#### 3.2 Economic analysis

Parameterized equations can estimate the equipment costs as a consistent technique for price analysis. The equations adopted for the costs of pumps associated with their electric motors, compressors, shell and tube exchangers, boilers, distillation towers, cooling towers, flash vessels, and water treatment systems can be found in Seider *et al.* (2016). To calculate the capital costs (CAPEX) it need to be considerate the equipment costs and general costs (CAXIANO *et al.*, 2020).

The total production cost includes all annual expenses for a chemical plant. For its calculation, expenses with utilities, water, electricity, and natural gas, and expenses with labor, raw materials, production costs, among others, are considered. For utility costs, the reference values are equal to \$0.0157/m3, \$18.72/GJ, and \$2.99/GJ for water, electricity, and natural gas, respectively (TURTON *et al.*, 2016). For labor expenses, 8000 hours of plant operation per year were considered, with three shifts of 5 workers per shift with annual payments of \$50,000 per operator. The other production expenses are described in Table 2 based on capital costs and labor expenses (LE).

Table 2: General heuristics of the total production cost.

Category	Parameters	Values
<b>Fixed costs (FC)</b>	maintenance	5% of CAPEX
	General costs	5% of LE
	Insurance, license fees and royalties	2% of CAPEX
<b>Variable costs (VC)</b>	PPE, cleaning supplies, chairs and accessories, etc..	1% of maintenance
	Sales and <i>marketing</i>	2% of FC + VC
<b>Others</b>	Human resources, accounting, financial etc.	35% of LE

#### 3.3 Indicator analysis

Eco-efficiency indicators or key performance indicators are essential parameters for planning and controlling industrial activities, allowing the establishment of transparent and reasoned analysis for decision-making (CAXIANO *et al.*, 2020; MANGILI *et al.*, 2019). These metrics can be divided into three categories: environmental, economic, and social, comprising the main aspects of the sustainability of a process (RUIZ-MERCADO *et al.*, 2012).

The present work will analyze three eco-efficiency indicators: two environmental indicators, water and CO2 emission, and one economic indicator, specific energy cost. For their comparison was used the Comparative Eco-efficiency Index (IEC) propose by Pereira *et al.* (2018).

## 4 Result and Discussions

The water consumption indicator was calculated and its results for each process are shown in Table 3.

The table results displayed that the P2 process performed the highest water consumption per quantity of product. The VRP1 intensification presented the best results for the indicator among the processes studied, followed by the VRP2 proposal. Therefore, vapor recompression intensification provides better results than conventional processes based on water consumption.

CO<sub>2</sub> emissions are associated with electricity consumption and natural gas burning. Table 4 lists all equipment associated with CO<sub>2</sub> emissions in each process and their respective energy consumption.

Table 3: Results for the water consumption indicator.

Streams	Consumption (m <sup>3</sup> /h)			
	P1	P2	VRP1	VRP2
<b>Makeup of cooling system</b>	43,46	54,44	12,16	16,17
<b>Makeup of steam system</b>	3,49	4,27	-	-
<b>Total consumption (m<sup>3</sup>/h)</b>	46,95	58,71	12,16	16,17
<b>Production (t<sub>product</sub>/h)</b>	11,67	11,67	11,67	11,67
<b>Indicator (m<sup>3</sup> H<sub>2</sub>O/t<sub>product</sub>)</b>	4,023	5,029	1,042	1,385

Table 4: Energy consumption by equipment in each process.

Energy consumption (GJ/h)			
P1	P2	VRP1	VRP2
46,5370	56,3772	8,9825	10,4877

The results for energy consumption showed a behavior similar to the water consumption, with the P2 process having the highest energy consumption of the plants, while the VRP1 process has the most economical energy consumption. Furthermore, the intensifications exhibited lower energy values than their conventional processes, corresponding to an energy saving of 80.70% of VRP1 compared to P1 and 81.40% of VRP2 compared to P2.

However, electricity consumption increased by 106.25% from P1 to VRP1 and 141.19% from P2 to VRP2. This increase happens due to adding a new compressor in the intensification processes, responsible for a significant increase in electrical energy demand.

The energy consumption were used to determine the CO<sub>2</sub> emissions applying their respective conversion factors. The results for CO<sub>2</sub> emissions are shown in Table 5.

Table 5: Results for the CO<sub>2</sub> emission indicator.

Data	Emission (t <sub>CO<sub>2</sub></sub> /h)			
	P1	P2	VRP1	VRP2
<b>Total emission (t<sub>CO<sub>2</sub></sub>/h)</b>	2,5075	3,0546	0,1877	0,2192
<b>Production (t<sub>product</sub>/h)</b>	11,67	11,67	11,67	11,67
<b>Indicator (t<sub>CO<sub>2</sub></sub>/t<sub>product</sub>)</b>	<b>0,2148</b>	<b>0,2617</b>	<b>0,0161</b>	<b>0,0188</b>

The energy cost indicator is calculated based on the results in Table 4 for electricity and natural gas costs and the total cost of operation. The final values of this indicator for each process are presented in Table 6.

The values shown in the table indicate that the VRP1 intensification has the lowest operating costs with energy, followed by the conventional P1 process. It is also noted that, despite the removal of natural gas costs, the intensification proposals considerably increase electricity costs due to the introduction of a new compressor.

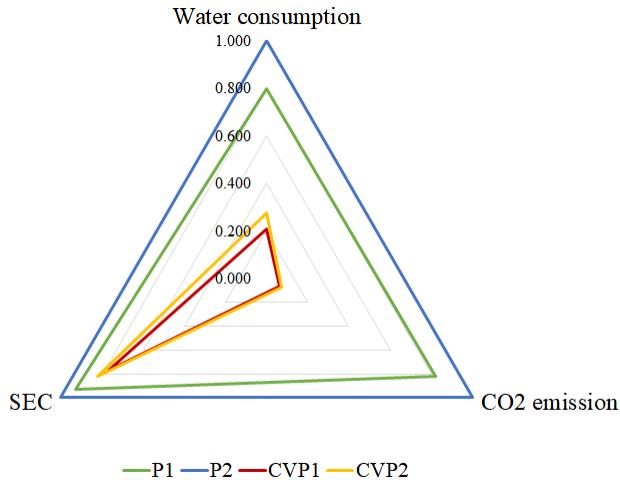
Finally, although the VRP1 and VRP2 processes present high operating costs, their results for the specific energy cost indicator were better than the conventional processes, indicating that the intensification reduces the fraction of operating costs related to energy consumption.

After normalization of each indicator, their values were plotted in a radar graphic, as shown in Fig 6.

Table 6: Results for the specific energy cost indicator.

Data	P1	P2	VRP1	VRP2
<b>Electricity (\$)</b>	\$426.348,41	\$446.822,83	\$1.305.702,87	\$1.524.496,53
<b>Natural gas (\$)</b>	\$1.043.005,78	\$1.275.013,81	-	-
<b>Energy cost (\$)</b>	\$1.469.354,19	\$1.721.836,65	\$1.305.702,87	\$1.524.496,53
<b>TPC (\$)</b>	\$3.419.033,89	\$3.729.155,62	\$3.684.712,39	\$4.009.206,04
<b>SEC (\$/\$)</b>	0,430	0,462	0,354	0,380

Figure 6: Process radar charts.



The graphic exhibited that the process P2 presents the largest area in the radar graph. The intensification proposals displayed a lower area for all indicators than conventional processes, with the most significant reductions observed in the CO<sub>2</sub> emission indicator. Finally, Table 7 presents the areas of the radar charts, calculated using the IEC method.

Table 7: Results for the Comparative Ecoefficiency Index.

Processes	P1	P2	VRP1	VRP2
<b>Area</b>	0.938	1.299	0.095	0.132
<b>IEC</b>	27.82%	-	92.70%	89.81%

The results presented in the table indicate that the P2 process presents the radar graph with the largest area, being, therefore, the least eco-efficient alternative to the indicators considered in the analysis. Likewise, analyzing the 27.82% eco-efficient gain of the P1 process, it is possible to state that,

among the conventional processes, separating propylene-propane mixture with two columns of 100 plates is a more eco-efficient alternative than using a column with 150 plates.

A considerable improvement in the plant eco-efficiency is observed regarding the intensified processes, reaching values close to or above 90%. Furthermore, it is possible to conclude from the IEC results that the VRP1 process presents the most significant gain in eco-efficiency, being 92.70% more eco-efficient than the P2 process, which is the worst alternative. Thus, it appears that the VRP1 process is the most eco-efficient alternative for the separation of the propylene-propane mixture among the studied alternatives to the three chosen indicators.

Applying the ICE methodology only to the VRP1 and VRP2 processes can verify that the VRP1 intensification is 28.56% more eco-efficient than the VRP2 technology, which was the second-best alternative among the studied processes. These results, in turn, demonstrate the considerable gain in eco-efficiency provided by the vapor recompression of the P1 process, despite both intensifications having presented similar results in the joint analysis of all alternatives.

## 5 Conclusion

In this work, the eco-efficiency analysis of two processes of distillation of the propylene-propane mixture present in the literature and their respective intensifications by vapor recompression was carried out. All processes were simulated in UniSim® software to obtain the necessary data for the calculation of eco-efficiency indicators. Also, in process P2, a storage system was added to allow a fairer analysis compared with process P1.

Concerning water consumption and CO<sub>2</sub> emissions indicators, the intensifications showed decreases in their conventional processes, conferring a reduction in water consumption of up to 72.02%. This reduction is justified by eliminating the reboilers, which removes the consumption of natural gas and reducing the condensers' thermal loads, which decreases the demand for water in the cooling systems.

On the last indicator, the specific energy cost, it is shown that the P2 process has the most significant value because of its high energy demand. Furthermore, the intensification proposals reduced the SEC compared to conventional processes.

Using the methodology of the Comparative Eco-efficiency Index, it was verified that the P1 process, the distillation with two columns of 100 plates, is the best alternative among conventional technologies, having a 27.82% gain in eco-efficiency compared to the P2 process, which uses only one column of 150. Moreover, it was found that the two intensification proposals presented eco-efficiency improvements of at least 90% with the P2 process, which was the worst alternative among those studied. The VRP1 process presented an eco-efficiency gain of 92.70%, being, therefore, the best alternative for separating the propene-propane mixture from the point of view of the analyzed indicators.

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