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Partial energy integration between biofuels production processes: Effect on costs, CO_2 emissions and process safety

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ABSTRACT

Energy integration is a tool which allows reducing the heating and cooling requirements for production processes. This is particularly important in the processes for production of biofuels, since such processes are expected to have low environmental impact, which can be achieved by reducing the need for steam and cooling water. It is common to perform energy integration by making use of all the available streams. This approach may allow reducing as much as possible utilities' requirements, but other indicators may be affected, such as capital costs, since the number of required equipment is increased. Thus, in this work the effect of performing partial integration is assessed, i.e., selecting only a few streams to perform the energy integration. The effect of increasing the number of integrated streams is assessed in terms of sustainability indicators based on the green chemistry principles. The studied indicators are utilities' requirements, total annual cost, environmental impact (assessed through CO_2 emissions) and safety (assessed through the HPSI index). The study is applied to the energy integration of a supercritical biodiesel production process and a lignocellulosic bioethanol production process.

1. Introduction

The production, transformation, and consumption of energy plays an important role in the challenge of climate change. These activities account for around two-thirds of global greenhouse gas emissions and about 90% of carbon dioxide emission, which is globally the most prevalent greenhouse gas (Lin et al., 2020). Efficient energy management in processing plants may lead to more profitability and more environmentally friendly industry. To achieve those goals, facilities with fluid material streams must consider heat exchange among these streams. Such a task is performed indirectly by heat exchangers. Proper allocation of these units includes selecting what streams are passing through them for a hot one to provide heat to a cold one. This mitigates the need for external utilities, which are also performed in heat exchangers identified as heaters or coolers. In heaters, a hot utility (e.g. steam from a boiler) is used to provide heat to a cold process stream; while in coolers, a cold utility (e.g. cold water from cooling towers) is employed as a cooling mean (Caballero et al., 2021). The set of heat exchangers implemented in a plant is named heat exchanger network (HEN). HEN synthesis is an intriguing area under the scope of industrial energy management. Moreover, as the number of streams in a plant grows, the number of possible stream matches becomes inconveniently large and turns out that exhaustively evaluating these combinations is computationally impossible with current technology. The development of strategies to overcome these complicating characteristics methodologically is thus of great value. A pioneering method is the noteworthy pinch analysis (Linnhoff and Flower, 1978; Flower and Linnhoff, 1980). Pinch analysis is very popular and successful because it is conceptually simple and with impressive results, i.e. 10–35% in energy savings. The pinch analysis principles are a set of rules, established using graphical representations such as Composite Curves, or by calculation-based methods known as the 'Problem Table Algorithm' (Flower and Linnhoff, 1980).

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Nomenclature

$C\%$ [%]	carbon content of the fuel
C_{inv} [USD]	capital cost
C_{op} [USD]	operational cost
$[CO_2]_{em}$ [kg/s]	emissions of carbon dioxide
EMR [kW·h/kg]	energy-to-mass indicator
EER [kW/kW]	delivered energy-to-required energy indicator
h_{proc} [kJ/kg]	enthalpy of steam
$I_{\Delta H_c}$ []	normalized score for heat of combustion
I_ρ []	normalized score for density
I_{FP} []	normalized score for flash point
I_{MF} []	normalized score for molar flowrate
I_P []	normalized score for pressure
$\dot{m}_i$ [kg/h]	mass flowrate of the product i
n [y]	payback period
NHV [kJ/kg]	net heating value of fuel
Q_{del} [kW]	heat delivered by the produced fuel
Q_{fuel} [kW]	fuel duty
Q_{proc} [kW]	process equipment duty
$Q_{proc,tot}$ [kW]	total heating requirements for the process
TAC [USD/y]	total annual cost
T_{FTB} [°C]	flame temperature of the boiler flue gases
T_{stack} [°C]	stack temperature
T_0 [°C]	ambient temperature
W []	empirical constant for the calculation of HPSI

Greek letters

λ_{proc} [kJ/kg]	latent heat of steam

On the other hand, the governmental restrictions on discharges of greenhouse gases and the steep changes in fossil fuel prices, shifted the worldwide trend to focus on renewable energy sources. Biodiesel and bioethanol are the most important liquid biofuels employed in the transportation sector due to their similarity with current crude oil-based fuels and their compatibility with current engines. Biodiesel, which contains mostly fatty acid methyl esters (FAME), is usually obtained from oils or fats via transesterification. Among the advantages of biodiesel, its similar properties (e.g. viscosity and volatility) compared to fossil diesel can be mentioned. Because of this, it can be used in standard diesel engines without requiring modifications (Kiss, 2010). Bioethanol is a liquid biofuel which can be produced from a large variety of natural renewable materials and conversion technologies. Due to its high octane and low cetane numbers and its high heat of vaporization, bioethanol is appropriate for blending with gasoline. Moreover, the production of bioethanol reduces the consumption of crude oil and the associated global greenhouse gas emissions. To become a viable alternative, biofuels should be economically competitive, show environmental benefits, and provide a high net energy gain (Severson et al., 2012).

The production of anhydrous bioethanol is very energy-demanding process, a major reason being the azeotropic distillation required to producing pure ethanol. Hence, various process integrations have been proposed to reduce the energy requirements. Since bioethanol can be used in the biodiesel production, many bioethanol plants integrate nowadays biodiesel production. Such an integrated bioethanol and biodiesel plant was developed by Dedini and it is being used since 2006 by Barralcool Mill, in State of Mato Grosso, Brazil (Kiss, 2010). Given the viability of operating biodiesel and bioethanol production plants in an integrated manner, and in order to make their operation economically profitable and generate fully sustainable processes, the possibility opens up to establish a conceptually operable, economically viable and environmentally friendly design in production of biofuels, through the application of heat exchanger networks (Brunet et al., 2015).

In the case of biodiesel production, it is important to develop process to treat low-cost vegetable oils, since the use of those oils may reduce the production cost of biodiesel by 60–70% (Farooq et al., 2015). One of the alternatives to treat those low-quality oils is by simultaneous esterification/transesterification with supercritical alcohols, as methanol or ethanol (Saka and Kusdiana, 2001; Demirbas, 2009). This kind of treatment has high energy requirements, mainly to take the reactants to the required reaction temperature. On the other hand, supercritical processes require less units, since less by-products are obtained.

The discussion around the sustainability of biomass and bioenergy use has begun in the previous decade in 2007 when the food versus energy debate was started due to the concurrent significant increase in energy and food prices. In terms of the "sustainable bioenergy" concept, the need to measure to which extend a biofuel or a production chain is sustainable is more than necessary. The environmental, social, and economic impacts of all phases (i.e. from land preparation and biomass feedstock collection to the final product distribution and consumption) must be carefully taken into consideration and be assessed based on well-defined sustainability indicators (Christoforou and Fokaides, 2019). To promote the sustainable production of biofuels, the interaction between biophysical, socio- economic, and governance drivers is deemed necessary (Florin et al., 2014).

The involved stakeholders in the production of biofuels need to pay attention on several aspects of sustainability that may arise. Selecting the most important metrics based only on greenhouse gases is a popular approach. To reduce the chance of burden shifting in space along the supply and demand networks and between different types of flows, green indexes should be incorporated into the process design if processes with sustainability and green chemistry characteristics are desired (Jiménez-González and Constable, 2011). According to Jiménez-González and Constable (2011), the indexes that contribute most to the concepts of sustainability and green chemistry are inherent safety, economy, and greenhouse gas emissions.

This work analyzes different topologies, based in energy integration, in an integrated biorefinery scheme where biomass is used to produce diverse biofuels. The analysis is performed in terms of various green indexes. To the authors knowledge, a study on the sustainability of the use of heat exchanger networks in integrated plants for the production of biodiesel and bioethanol has not been reported, nor has a sustainability analysis has been reported for different degrees of energy integration in heat exchanger networks. Among the works analyzing the effects of energy integration in the production of biofuels, the report by Chouinard-Dussault et al. (2011) can be mentioned, where the energy integration for a set of case studies is analyzed, including a process for bioethanol production and a biodiesel production process, assessing the effect of the energy integration in a life cycle analysis frame. Brunet et al. (2015) reported the energy integration of an alkali-catalyzed biodiesel production process, reporting reduction of 3.19% for production cost and 9.31% for energy requirements. In the same work, the energy integration of a bioethanol production process is performed, obtaining savings of 1.71% in cost and 7.13% for energy requirements. Petersen et al. (2015) reported the energy integration of two routes to produce bioethanol: hydrolysis followed by fermentation, and simultaneous saccharification and fermentation. Gutiérrez-Antonio et al. (2016) reported the energy integration of conventional and intensified processes to produce biojet fuel by hydrotreating of *Jatropha curcas* oil, obtaining savings in utilities costs up to approximately 35% in comparison with the conventional, non-integrated process. Most of the reported studies related to the energy integration in biofuels production processes perform the integration of all the streams releasing/requiring energy, measuring the advantages of energy integration in terms of the utilities savings. Nevertheless, other indicators as environmental impact and process safety are usually not assessed. To ensure the sustainability of a biomass conversion process, a variety of indicators must be evaluated, including environmental, economic and social indicators (Van Schoubroeck et al., 2018). Such measurements can give insights about how the process accomplishes with the green chemistry principles (Abdussalam-Mohammed et al., 2020). In the same line, Jiménez-González et al. (2012) mentioned the necessity of assessing "green metrics" when designing a process to achieve a broader target of sustainability. Among those metrics the aspects of environmental, economics and inherent safety can be highlighted. Further, modification in the topology, due to energy integration, for a given process can also modify sustainability indexes (Morseletto, 2020; El-Halwagi and Foo, 2021). This work aims to fill this gap in the area of integrated bioprocesses. In a recent work, the effect of partial energy integration is studied for an ethylene/propylene production process, evaluating the effect of such kind of integration through the equipment and utilities costs, the return of investment (ROI), the emissions of CO_2, and the Inherent Safety Index for Shell and Tube Heat Exchangers (ISISTHE). According to the reported results, the partial integration may allow a compromise between the analyzed indicators (Ortiz-Espinoza et al., 2020). The use of ISISTHE could not be appropriate for analyzing biofuel processes since it is defined only to evaluate chemical processes and explosion scenarios (Pasha et al., 2017).

To perform the energy integration, the pinch point methodology is used in this proposal. Additionally, different scenarios with partial energy integration are analyzed, implying that only some streams are included in the pinch analysis. The effect of such partial integrations is assessed in terms of total energy requirements, total annual cost, environmental impact and process safety, comparing the obtained results with the total integration. Environmental impact is measured in terms of CO_2 emissions, energy required per kilogram of products, and the energy delivered/energy required ratio. On the other hand, process safety is measured in terms of the HPSI and the relative risk (López-Molina et al., 2020a). It is important to mention that the HPSI has not been previously used to assess the safety on integrated biorefinery schemes. Its previous use has been focused on single biofuels production processes. Following the classification given by Van Schoubroeck et al. (2018), the energy requirements and the CO_2 emissions represent the environmental indicators, the total annual cost is the economic indicator, and the HPSI represents the social indicators.

2. Case study

Two biofuels' production processes are analyzed in this work. The first one implies the production of biodiesel from waste cooking oil by simultaneous transesterification and esterification using supercritical ethanol (Fig. 1). The second one is referred to the conversion of lignocellulosic biomass into bioethanol (Fig. 2). These processes have been previously reported by Gómez-Castro et al. (2017) and Aldana-González et al. (2022).

In the biodiesel production process, the oil and ethanol streams are conditioned before entering to the reactor, where the triglycerides are transesterified and the free fatty acids are esterified. Next, the pressure is reduced to 1 bar and the stream goes through a purification step to recover the ethanol and the glycerol, while the biodiesel is obtained with the desired purity. In the bioethanol production process, biomass is first pretreated with sulfuric acid to remove the lignin, then hydrolyzed to obtain sugars from the cellulose and hemicellulose. Finally, the sugars are fermented to produce bioethanol. Excess water and by-products are removed in a distillation train, where the ethanol-water azeotrope is overcome by using an extractive distillation system with glycerol as extractant.

The streams requiring for cooling or heating are highlighted in Figs. 1 and 2. Fig. 3 shows the streams diagram, while Table 1 presents the properties for all the streams to be integrated. The information shown in Table 1 has been taken from simulations of the processes, performed in Aspen Plus V. 8.0. The processes involve nine cold streams and nine hot streams with a wide range of temperatures, between 25 and almost 475 ºC.

To evaluate the effect of partial integration, several scenarios have been studied:

Scenario I: All the streams are integrated.

Scenario II: The streams are integrated only inside the process to which they belong, i.e., streams C2, C4 and C8 are integrated with streams H4, H6, H8 and H9; while streams C1, C3, C5, C6, C7 and C9 are integrated with streams H1, H2, H3, H5 and H7.

Scenario III: The three coolest streams requiring heating are integrated with the three hottest streams requiring cooling, i.e., streams C1, C2 and C3 are integrated with streams H7, H8 and H9. This scenario would represent an integration of approximately 33% of the streams.

Scenario IV: The five coolest streams requiring heating are integrated with the five hottest streams requiring cooling, i.e., streams C1, C2, C3, C4 and C5 are integrated with streams H5, H6, H7, H8 and H9. This scenario would represent an integration of approximately 55% of the streams.

Scenario V: The eight coolest streams requiring heating are integrated with the eight hottest streams requiring cooling, i.e., streams C1, C2, C3, C4, C5, C6, C7 and C8 are integrated with streams H2, H3, H4, H5, H6, H7, H8 and H9. This scenario would represent an integration of approximately 89% of the streams.

It can be observed that the number of streams to be integrated gradually increases from scenarios III to V. It has been decided to integrate the hottest streams with the coolest streams to ensure the existence of potential exchanges in terms of the temperature levels for each kind of stream. All the scenarios are compared with the case where no integration takes place.

3. Methodology

3.1. Energy integration

Using the information available for the streams in both processes, the energy integration is performed through the pinch point methodology (Linnhoff and Hindmarsh, 1983). Only the streams involved in each scenario are considered to perform such study. The heating/cooling requirements for the streams not included on a given scenario are satisfied by either cooling water or steam. For each scenario, a heat cascade is developed for the streams to be integrated, using $\Delta T = 10\,^{\circ}C$. From the heat cascade, the pinch point and the minimal heating and cooling requirements are determined. Using this information, feasible exchanges are defined between the hot and cold streams analyzed on each case. Those exchanges must accomplish the following constraints (Linnhoff and Hindmarsh, 1983):

1. There must be no streams crossing the pinch point.
2. There must exist no temperature crossing on the exchangers
3. The minimal number of exchangers must be accomplished in both sides of the pinch point

Once each network is completed, it is verified that the network accomplishes the heating and cooling minimal requirements predicted by the heat cascade, and the calculation of the indicators is performed.

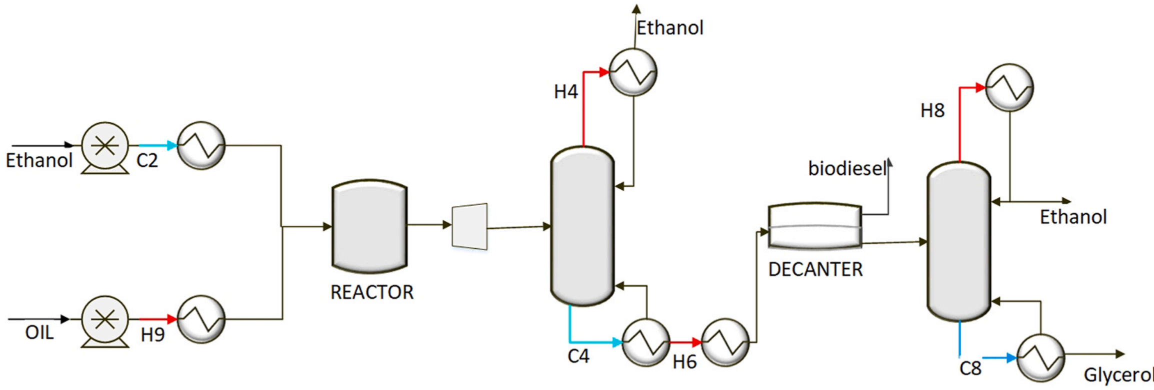

Fig. 1. Biodiesel production process with supercritical ethanol.

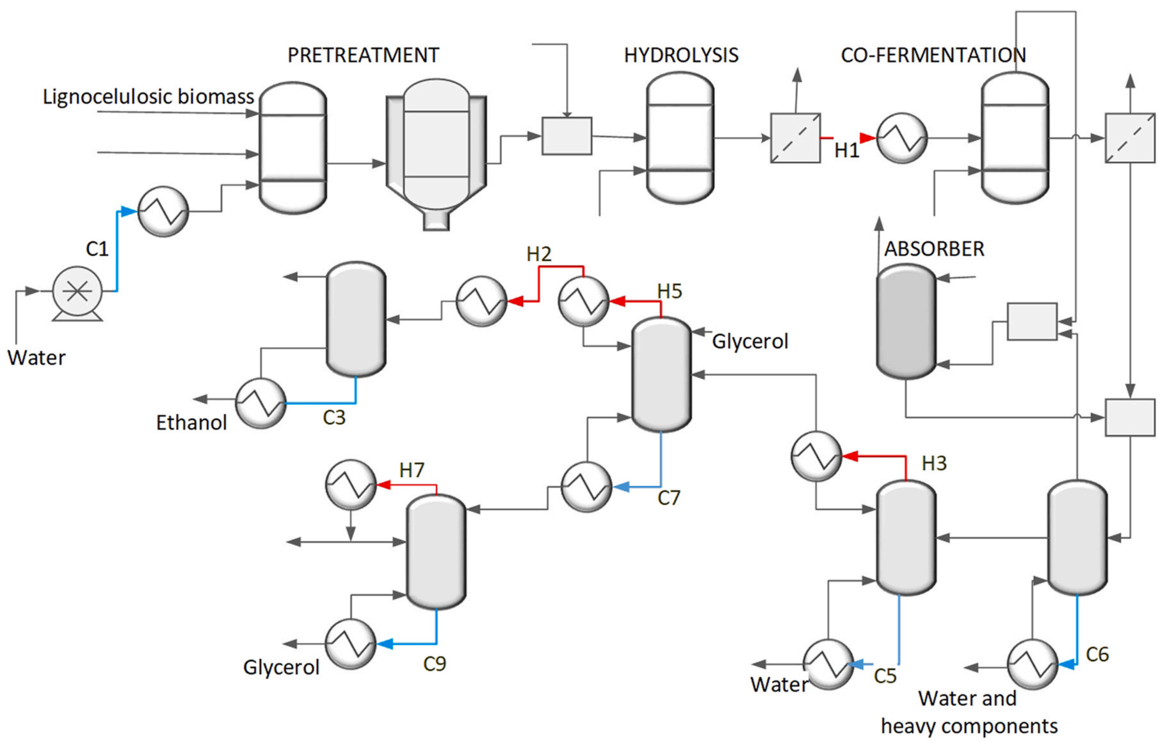

Fig. 2. Lignocellulosic bioethanol production process.

3.2. Energetic assessment

The energetic assessment is performed in terms of the heating and cooling required after integration took place. For scenarios I and II, such requirements are needed to accomplish the network's heating or cooling tasks. For the scenarios where only partial integration occurs (III, IV and V), these requirements are those needed to complete the heating or cooling tasks in the network, plus those needed to satisfy the heating/cooling requirements for the streams not considered in the integration network.

3.3. Economic assessment

Economic assessment has been performed by calculating the total annual cost (TAC) for the heating/cooling devices. TAC is given by:

$$TAC = \frac{C_{inv}}{n} + C_{op} \quad (1)$$

Where C_{inv} is the capital cost, given by equipment, and C_{op} is the operational cost, represented by the utilities' costs. n is the payback period, which in this work has been assumed as 5 years (Turton et al., 2012). Capital costs are estimated with Guthrie's method, as reported by Turton et al. (2012), updating the costs to 2018 with the corresponding Chemical Engineering Cost Index, which has a value of 556.8. Operational costs are estimated in terms of the utilities required for the process, i.e. steam and water. Electricity costs are not included since they are not affected by the energy integration. Unitary costs for utilities are shown in Table 2.

3.4. Environmental assessment

Environmental impact and sustainability of the proposal have been assessed in terms of three indicators. The first one is given by the emissions of carbon dioxide due to the production of the steam needed to fulfill the heating requirements of the process. Those emissions are estimated following the procedure reported by

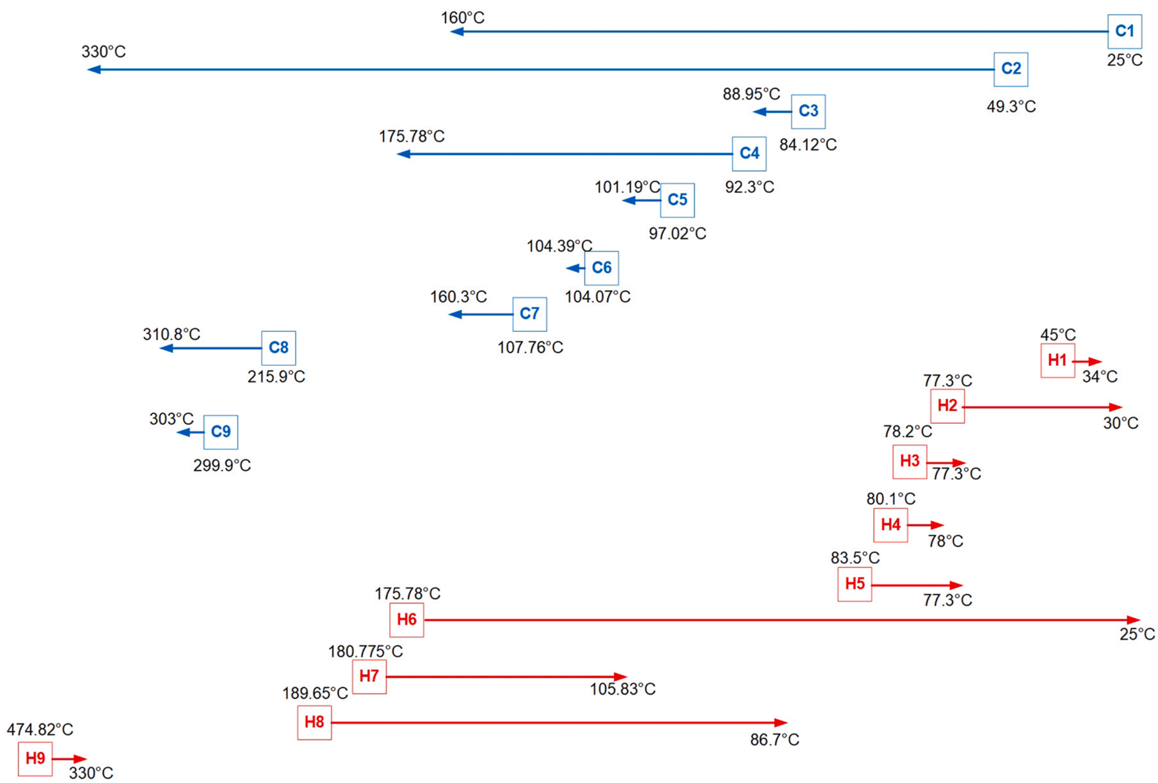

Fig. 3. Streams diagram for the biofuels production processes.

Table 1
Characteristics of the streams (C for cold streams, H for hot streams).

Stream ID	Description	Initial Temperature (°C)	Target Temperature (°C)	w*Cp (kW/°C)
BIODIESEL PROCESS				
C2	Ethanol for heating	49.3	330	3.82
C4	Bottoms of the column DD-1	92.3	175.78	5.16
C8	Bottoms of the column DD-2	215.9	310.8	0.3
H4	Top of the column DD-1	80.1	78	357.7
H6	Stream entering the decanter DECD-1	175.78	25	0.7
H8	Top of the column DD-2	189.65	86.7	0.035
H9	Oil for cooling	474.82	330	0.96
BIOETHANOL PROCESS				
C1	Water entering pretreatment reactor	25	160	45.46
C3	Bottoms of the column DE-4	84.118	88.95	38.84
C5	Bottoms of the column DE-2	97.017	101.19	705.2
C6	Bottoms of the column DE-1	104.067	104.387	25,249
C7	Bottoms of the column DE-3	107.76	160.3	15.28
C9	Bottoms of the column DE-5	299.9	303	351.3
H1	Stream entering co-fermentation reactor	45	34	67
H2	Stream entering DE-4	77.3	30	20.1
H3	Top of the column DE-2	78.2	77.3	5516.6
H5	Top of the column DE-3	83.5	77.3	108.2
H7	Top of the column DE-5	180.775	105.828	10.56

Table 2
Unitary utilities costs used for TAC estimation.

Utility	Unitary cost (USD/kg)	Source
Cooling water @20 ºC	0.0000148	Turton et al. (2012)
Cooling water @10 ºC	0.0000655	Sinnott (1993)
Steam @15 bar	0.02959	Turton et al. (2012)
Steam @150 bar	0.03054	Turton et al. (2012)

Gadalla et al. (2006), which is described next. Emissions of carbon dioxide, $[CO_2]_{em}$, are computed through Eq. (2):

$$[CO_2]_{em} = 3.67\left(\frac{Q_{fuel}}{NHV}\right)\left(\frac{C\%}{100}\right) \tag{2}$$

where Q_{fuel} (kW) is the duty to be delivered by a fuel used to produce steam, *NHV* (kJ/kg) is the fuel's net heating value, and *C%* is the carbon content of the fuel. The heat duty delivered by the fuel is given by:

$$Q_{fuel} = \frac{Q_{proc}}{\lambda_{proc}}(h_{proc} - 419)\frac{T_{FTB} - T_0}{T_{FTB} - T_{stack}} \tag{3}$$

where λ_{proc} is the latent heat of the steam, while h_{proc} is its enthalpy. Q_{proc} is the heat needed in the process equipment. T_{FTB} is the flame temperature of the boiler flue gases, T_{stack} is the stack temperature, and T_0 is the room temperature. Values of the parameters required to estimate carbon dioxide emissions are summarized in Table 3, assuming that natural gas is used as fuel.

Table 3
Values for the parameters used to estimate the emissions of carbon dioxide.

Parameter	Value	Units
Net heating value	51,600	kJ/kg
Carbon content	75.4	%
Flame temperature	1800	ºC
Stack temperature	160	ºC
Room temperature	25	ºC

The second indicator measures the ratio between the energy required for the process and the produced mass of fuel, i.e.:

$$EMR = \frac{Q_{proc,tot}}{\dot{m}_i} \quad (4)$$

where EMR is the energy-to-mass indicator, $Q_{proc,tot}$ is the total heating requirements for the process, and $\dot{m}_i$ is the mass flowrate of the product *i*. This indicator is estimated (i) only for the biodiesel process, (ii) only for the bioethanol process, and (iii) for both processes, implying the mass flowrate of the two main products.

The third indicator is given by the ratio between the energy delivered by one of the obtained fuels and the energy required to produce it, i.e.:

$$EER = \frac{Q_{del}}{Q_{proc,tot}} \quad (5)$$

where EER is the delivered energy/required energy ratio and Q_{del} is the heat delivered by the produced fuel, either bioethanol or biodiesel. To estimate the delivered energy, the combustion heat of bioethanol (27,729.54 kJ/kg) and biodiesel (36,709.89 kJ/kg) is used, along with their respective mass flow rates.

3.5. Risk assessment

The effect of partial energy integration on the safety of biodiesel and bioethanol processes was evaluated using two strategies, the identification of hazardous streams and the relative risk of the process. This evaluation intended to identify the number of dangerous process streams for each scenario described in the case study, since the main effect that energy integration has on the process is the increase in the number of process streams resulting from incorporating additional heat exchangers. These streams can either be dangerous or not and may come from inside the process or from outside the process. Therefore, the number of heat exchanges, the number of hazardous streams, the number of hazardous shared streams, and the relative risk are evaluated for each process and each scenario. With these parameters, it is observed how the safety of each process is affected by the level of energy integration. The number of dangerous process streams is determined using the HPSI (López-Molina et al., 2020a). This index involves the flash point, the heat of combustion, the density, the molar flow, and the pressure of each stream to define its hazard. The HPSI is defined as follows:

$$HPSI = \left(\frac{I_P \cdot I_{MF} \cdot I_{\Delta H_c} \cdot I_{FP}}{I_\rho}\right) \cdot W \quad (6)$$

where I_P, I_{MF}, $I_{\Delta H_c}$, I_{FP} and I_ρ are normalized scores for the pressure, molar flowrate, heat of combustion, flash point and density, respectively; W is an empirical constant dependent on the magnitude order desired for the results. All the data required for the calculation of HPSI were obtained from the Aspen Plus simulation. Once the HPSI was evaluated, the relative risk of each scenario was determined for the biodiesel and bioethanol process using the strategy proposed by López-Molina et al. (2020b), which normalizes the HPSI values of the streams to assign a level of risk to the process. The risk of each process will change based on these two indicators and the number of hazardous process streams. This information helps finding which scenarios are the safest.

4. Results

4.1. Process integration

Once the heat cascades have been developed, the following pinch points are detected:

Scenario I: 73.5 ºC for cold streams, 83.5 ºC for hot streams.

Scenario II (biodiesel process): 70.1 ºC for cold streams, 80.1 ºC for hot streams.

Scenario II (bioethanol process): 73.5 ºC for cold streams, 83.5 ºC for hot streams.

Scenario III: 25 ºC for cold streams, 35 ºC for hot streams.

Scenario IV: 25 ºC for cold streams, 35 ºC for hot streams.

Scenario V: 73.5 ºC for cold streams, 83.5 ºC for hot streams.

It is observed that the pinch points for scenarios I, II and V are similar, while there is an important modification for scenarios III and IV, since only the colder and hotter streams are involved in the analysis. For such cases, all the cold streams are located to the left of the pinch point. This can be observed in the streams diagram for each network, which are shown in Figs. 4–8. For scenario I, it is interesting to observe that even if all the streams for both processes are included in the pinch analysis, there is only a single exchange between streams of different processes, i.e. stream C2 of the biodiesel process with stream H5 of the bioethanol process. All the other streams are integrated inside their own process, which makes the obtained network like that of scenario II.

For scenario III, only three cold streams and three hot streams are included in the pinch analysis. Nevertheless, this configuration is enough to fully satisfy the cooling requirements, as observed in Fig. 6. A similar observation can be made for scenario IV, where all the cooling requirements for the 5 hotter streams are fulfilled.

4.2. Energetic assessment

Table 4 summarizes the heating and cooling requirements for each of the analyzed scenarios. For the scenario with no integration, it is observed that the total cooling requirements are considerably high. As partial integration occurs, the heating requirements are gradually reduced from 4.49% to almost 16%. On the other hand, if only three hot and three cold streams are included in the energy integration, cooling requirements are only slightly reduced. On the scenario where five cold and five hot streams are integrated, the cooling requirements are greatly reduced, up to almost 91% for the fully integrated scenarios.

It is observed that there is only a slight difference between the heating and cooling requirements for scenario I and II. This can be explained in terms of the small interaction between both processes, for scenario I there is only a single exchange between one stream of the biodiesel process and one stream of the bioethanol process.

4.3. Economic assessment

Results for the economic assessment are presented on Table 5. As the number of streams involved on the energy integration increases, more savings are obtained in utilities costs. For scenario III, only 4.6% of utilities cost is reduced. On the other hand, the highest reduction for utilities cost is 15.7%, allowed by the total integration (scenario I).

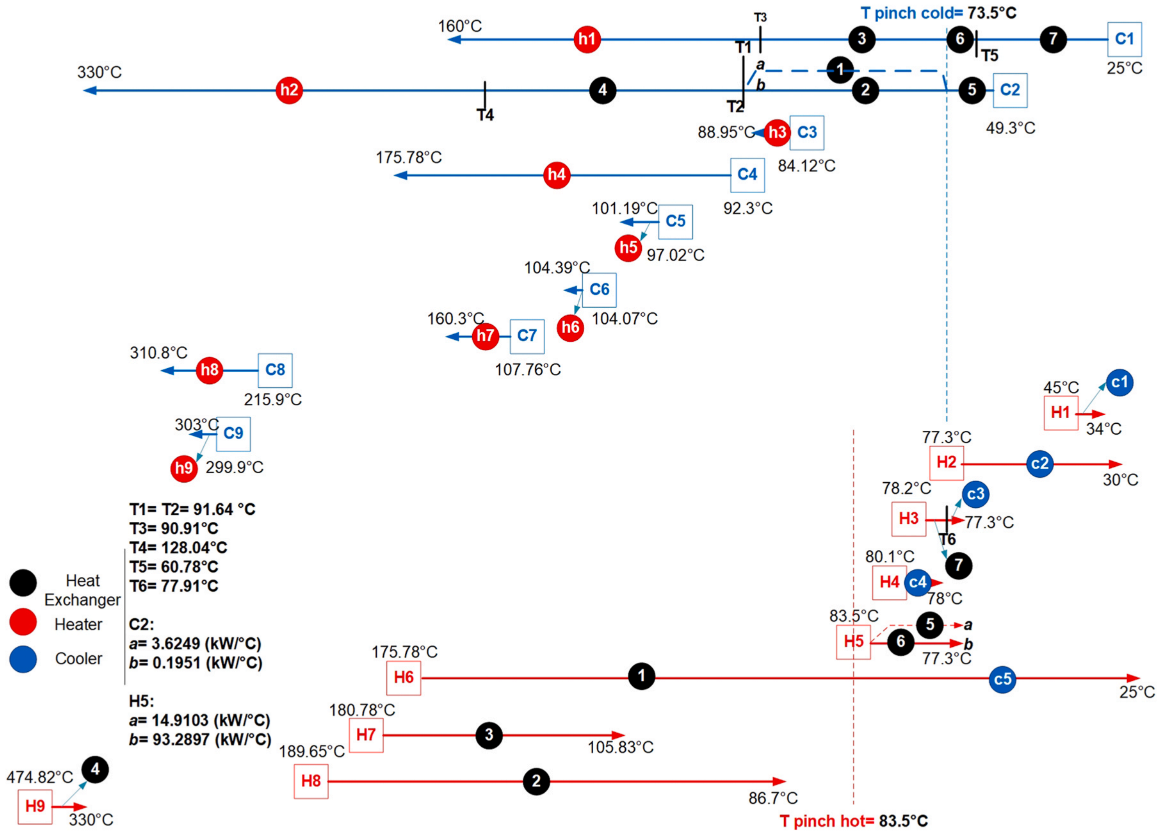

Fig. 4. Integrated network, scenario I.

It is observed that equipment cost increases for scenarios III, IV and V due to the need for additional exchangers. Nevertheless, as more streams are included in the analysis, the increment is lower. For scenarios I and II, there is even a reduction in the equipment cost. This is explained in terms of the steam required to satisfy the heating requirements of the high-temperature streams. When no integration or only partial integration occurs, heating requirements for such streams are satisfied with high-pressure steam, implying the need for more expensive materials to withstand the pressure levels. When energy integration occurs, such heating needs can be fulfilled with process streams at high temperature, but lower pressure. This implies the need for less expensive materials, which reduces the equipment cost.

4.4. Environmental assessment

Computed CO_2 emissions are presented on Table 6. It is observed that the bioethanol process contributes with approximately 90% of the emissions for the non-integrated case. Nevertheless, it is important to mention that this process is designed to produce approximately 3358 kg/h of bioethanol, while the production rate of biodiesel is 1412 kg/h. Even with this consideration, it is observed that the normalized emissions of CO_2 per kg of product are almost 3.5 times higher for the bioethanol production.

In terms of the energy integration, it is observed that the total emissions are slightly reduced when partial integration occurs, reaching a reduction of 15.71% when all the streams are included in the analysis. A similar trend is observed for the normalized emissions, where savings around 15% and 22% are obtained for the bioethanol and biodiesel production, respectively, when fully integrating the processes.

The sustainability indicators in terms of the energy use are shown in Table 7. It is clearly observed that, even on a normalized basis, the production of bioethanol requires higher energy input per kilogram of product. This can be explained in terms of the low efficiency of the conversion steps and the difficulty to separate the ethanol/water azeotrope. As integration takes place, the energy requirements are reduced, allowing reductions for this indicator of 15.7% and 19.4% for the bioethanol and biodiesel processes, respectively, when full integration takes place. In terms of the delivered energy, for the non-integrated case the potential delivered energy of bioethanol is only slightly higher than the energy required for its production, but this indicator can be enhanced through the energy integration. On the other hand, biodiesel can deliver much more energy than the required for its production, even in the non-integrated case. Through process integration, this indicator is considerably enhanced for biodiesel production.

In general terms, it has been observed that the bioethanol process is more energy-intensive than the supercritical biodiesel process. Moreover, in terms of the percentual savings, the biodiesel process is the most benefited with the application of the process integration techniques, in terms of energy use and CO_2 emissions.

4.5. Risk assessment

The safety indicators for the biodiesel process are presented in Table 8. When energy integration occurs, the number of dangerous process streams increases from 4 to 12 hazardous currents.

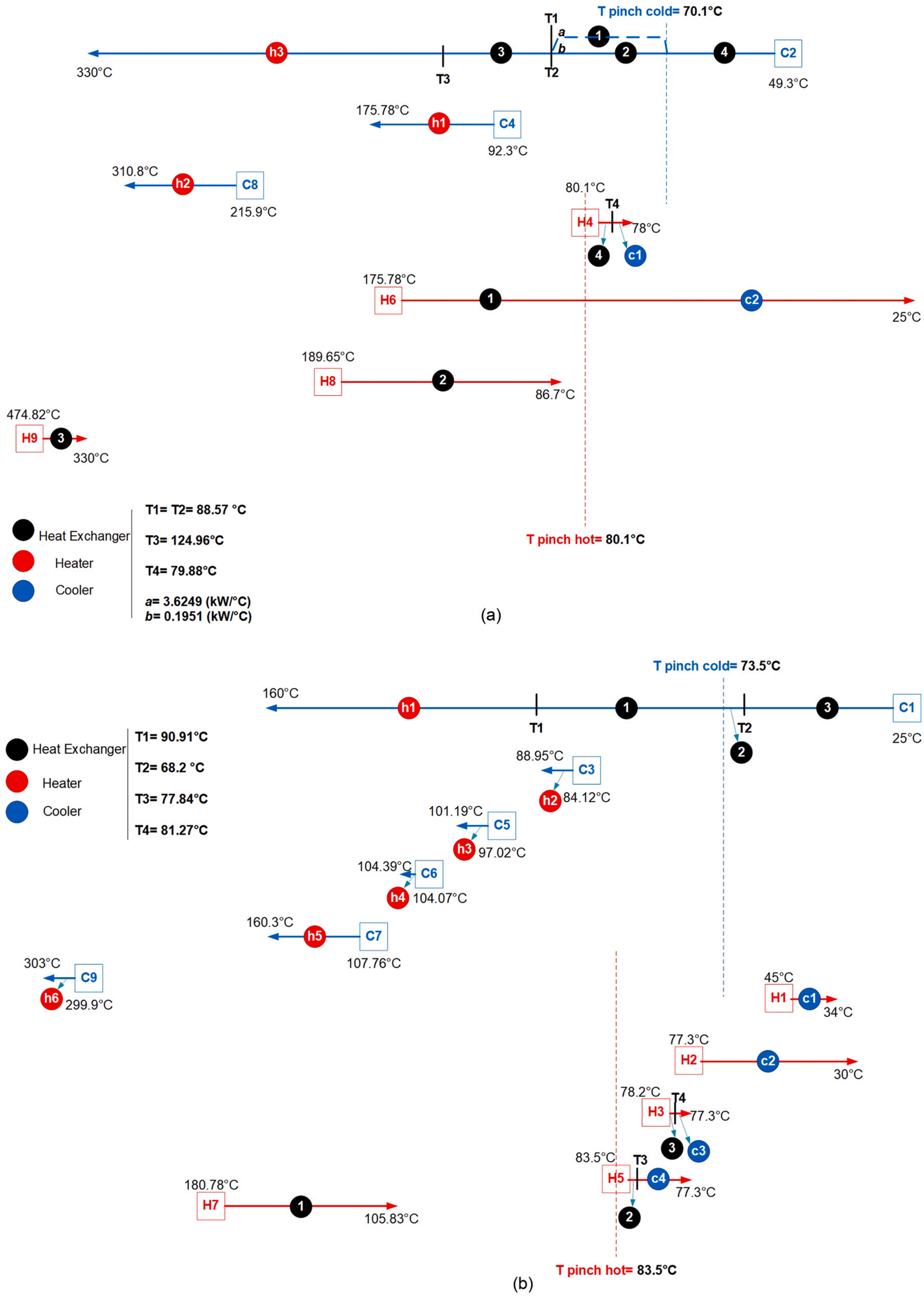

Fig. 5. Integrated network, scenario II (a) biodiesel process, (b) bioethanol process.

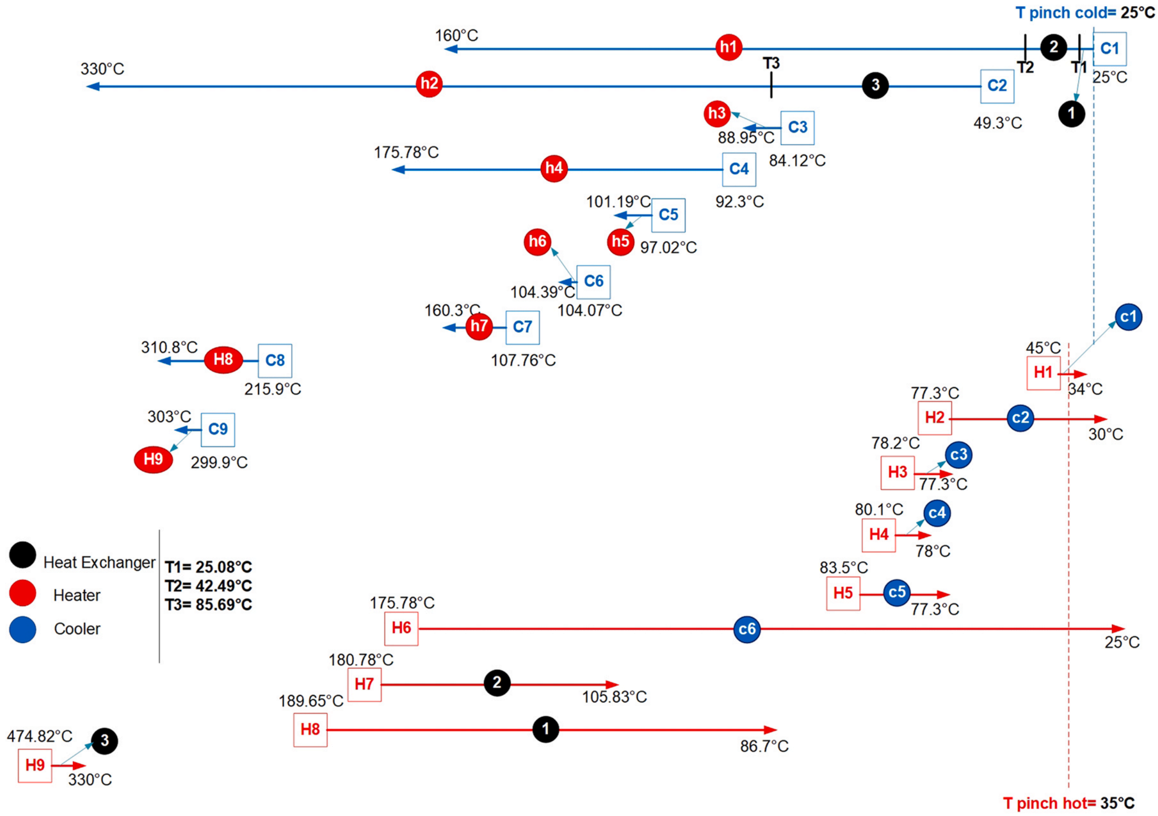

Fig. 6. Integrated network, scenario III.

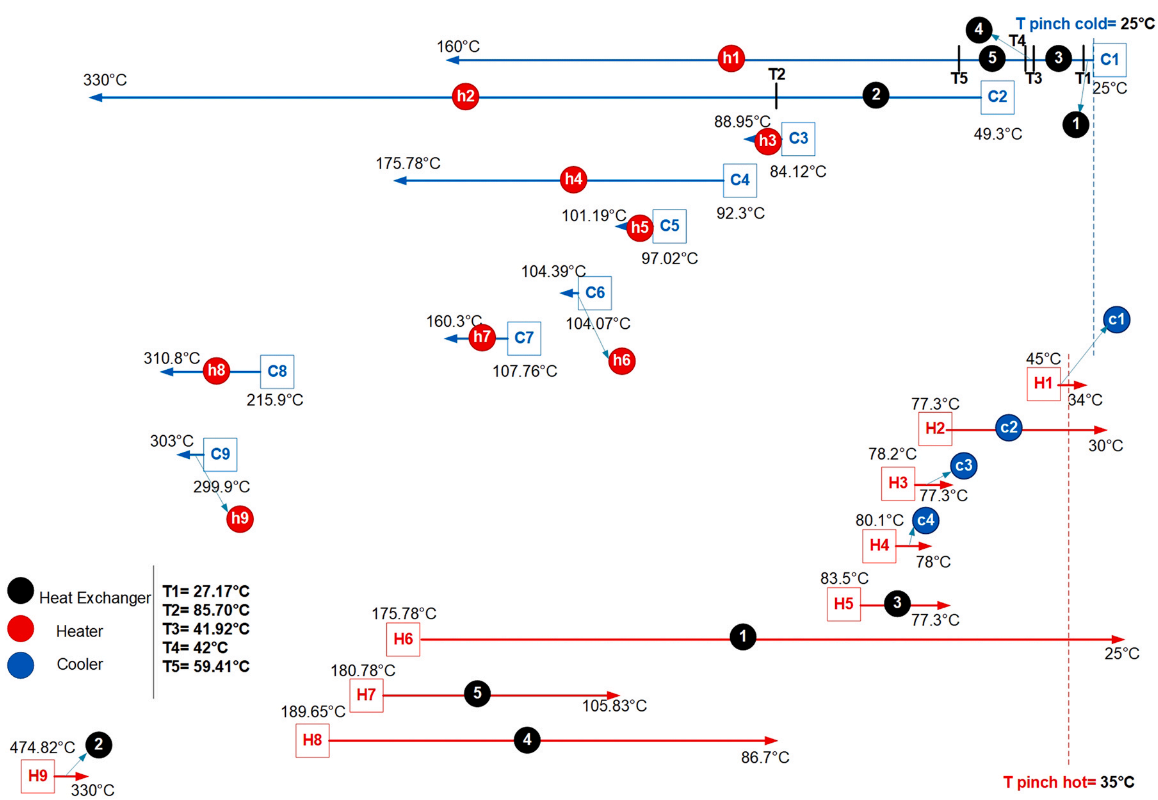

Fig. 7. Integrated network, scenario IV.

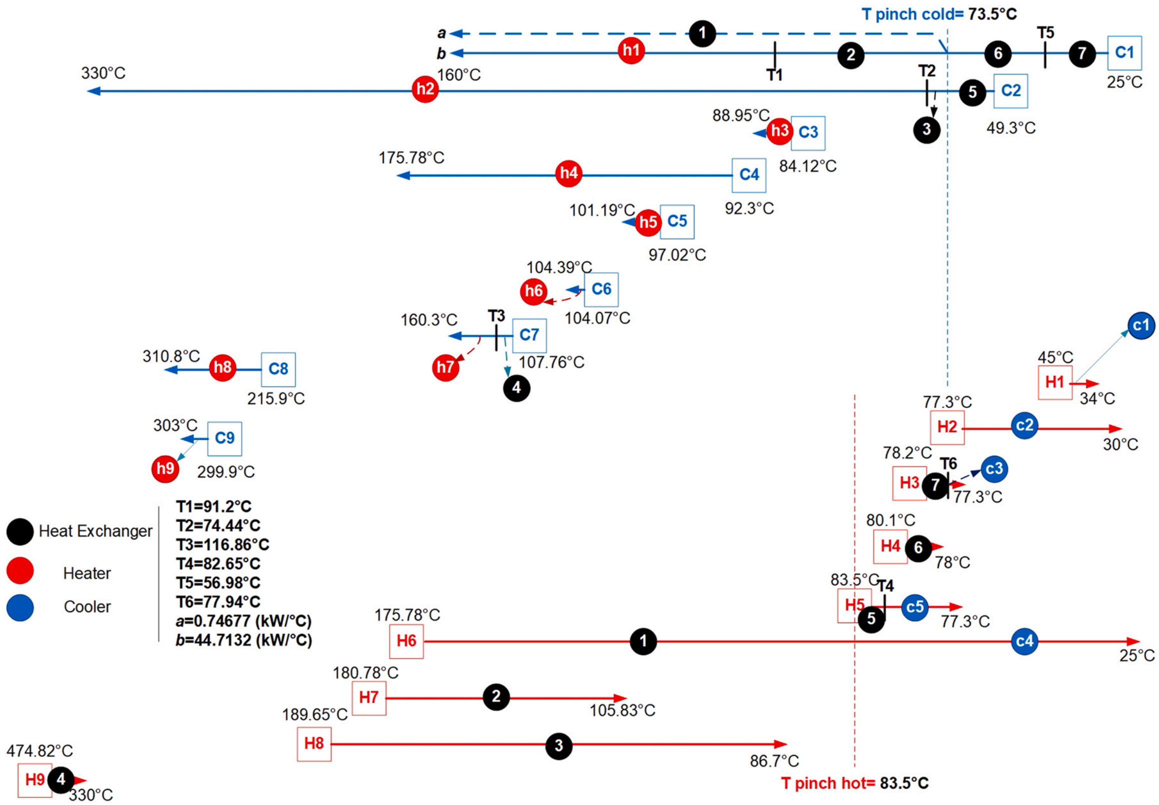

Fig. 8. Integrated network, scenario V.

Table 4
Heating and cooling requirements for each scenario.

Scenario	Heating (kW)	Cooling (kW)	Heating savings (%)	Cooling savings (%)
No integration	20,770.59	62,942.76	0.0	0.0
I	17,473.57	5818.49	15.87	90.76
II	17,485.31	5828.95	15.81	90.74
III	19,836.49	62,008.69	4.49	1.48
IV	19,067.14	7410.84	8.20	88.23
V	17,474.70	5957.46	15.86	90.53

Table 5
Results for the economic assessment.

Scenario	Equipment cost (USD)	Utilities cost (USD/y)	TAC (USD/y)	TAC savings (%)
No integration	4793,440.94	10,690,576.28	11,649,264.50	0.0
I	4373,083.63	9014,332.45	9888,949.18	15.11
II	4543,418.59	9025,302.46	9933,986.17	14.72
III	5775,925.65	10,190,863.91	11,346,049.04	2.60
IV	5378,950.99	9831,504.90	10,907,295.10	6.37
V	4831,762.14	9110,812.48	10,077,164.91	13.49

Table 6
Results for emissions calculation.

Scenario	Bioethanol process (kg/h)	Biodiesel process (kg/h)	Total emissions (kg/h)	Emissions reduction (%)
No integration	5120.43	613.47	5733.90	0.0
I	4357.42	475.81	4833.23	15.71
II	4357.42	481.19	4838.61	15.61
III	4917.96	549.85	5467.81	4.64
IV	4722.04	549.84	5271.88	8.06
V	4305.57	569.52	4875.09	14.98
Scenario	**Bioethanol process (kg CO_2/kg bioethanol)**	**Biodiesel process (kg CO_2/kg biodiesel)**	**Reduction on bioethanol process (%)**	**Reduction on biodiesel process (%)**
No integration	1.52	0.43	0.0	0.0
I	1.29	0.33	15.13	22.44
II	1.29	0.34	15.13	21.56
III	1.46	0.38	3.95	10.37
IV	1.41	0.38	7.78	10.37
V	1.29	0.40	15.13	6.97

The increase in dangerous currents is caused by the incorporation of new heat exchangers among hazardous process streams, which generate new arrangements, additional connections, and, consequently, increase the number of process lines. On the other hand, biodiesel processes' relative risk, either with or without integration, does not undergo significant changes, being at a low-risk level. A similar effect is observed in the bioethanol process, reported in Table 9. In this case, the number of dangerous currents without integration is seven and increases to ten when integrated with the biodiesel process. The relative risk in this process does not undergo considerable changes, remaining at a relatively low-risk level. In both processes, the number of dangerous process lines increases because the inherently hazardous currents of the process are used to integrate energy, increasing the number of equipment and pipes that transport them. The increase of dangerous lines and the different process conditions (pressure and density) increase the relative risk when both processes are either partially or fully integrated. Therefore, if it is desired to reduce the

Table 7
Sustainability indicators.

EMR			
Scenario	Bioethanol process (kW·h/kg bioethanol)	Biodiesel process (kW·h/ kg biodiesel)	Both processes (kW·h/kg biofuels)
No integration	5.73	1.08	4.35
I	4.83	0.87	3.66
II	4.83	0.88	3.67
III	5.49	0.98	4.16
IV	5.26	0.98	3.99
V	4.77	1.02	3.66

EER		
Scenario	Bioethanol process (kW bioethanol/kW)	Biodiesel process (kW biodiesel/kW)
No integration	1.34	9.40
I	1.59	11.69
II	1.59	11.58
III	1.40	10.34
IV	1.46	10.34
V	1.61	10.03

danger of the exchange network., the least dangerous streams must be the firsts chosen to be integrated. That is, it would be recommended to use as much as possible the least dangerous currents in energy integration.

Additionally, it is observed that some dangerous process streams remain in the vapor phase in various exchanges. Maintaining this phase in various equipment substantially increases the danger for the network since the release of dangerous substances in the vapor phase increases the process' risk significantly. Therefore, it is advisable to condense dangerous currents in the vapor phase as soon as possible. Finally, the biodiesel and bioethanol processes operate at significantly different pressures. This differential pressure can generate the collapse of pipes or equipment when integrating both processes. Thus, it is important to condition the streams to preferably low-pressure conditions to reduce equipment collapse scenarios and sudden substances expansion in emission cases.

4.6. Summary of results

According to the obtained results, as the number of streams used for energy integration increases, the savings on utilities significantly increases, up to almost 16% for heating and almost 91% for cooling. It has been determined that either integrating both processes or integrating each process as a separate entity, utilities' savings are almost the same. This occurs since there is only one feasible inter-process exchange. In terms of the economic analysis, as more streams are integrated, the TAC reduces in a higher proportion, up to approximately 15% for scenario I, where all the streams are integrated. Interestingly, for the scenarios I and II, there is a reduction on the capital cost compared to the scenario with no integration, since the need for high-pressure steam is avoided, thus materials with lower cost can be used for the exchangers. As more steams are integrated, CO_2 emissions are reduced, as could be expected due to the reduction on heating requirements. Additionally, better use of the energy is observed as the number of integrated streams increases. It is important to notice that the reduction on CO_2 emissions is strongly dependent on the selection of the streams to be integrated. With these indicators, it seems that the best alternative is to integrate the processes thoroughly. Nevertheless, the safety analysis indicates that, as more streams are integrated, the number of hazardous streams increases, with a higher number for the scenario where both processes are integrated. However, the scenario where the streams are integrated only inside their own process shows the highest relative risk.

Fig. 9 summarizes the obtained results. Some of the indexes have been scaled to simultaneously visualize the effect of integration on all the studied criteria. The most important effect of energy integration is observed in the cooling requirements. Nevertheless, this effect only occurs when integration occurs in more than 50% of the streams. It is observed that, as more streams are integrated, the TAC, the CO_2 emissions and the EMR are reduced, showing the positive effect of energy integration on those indicators. On the other hand, as the number of integrated streams is increased, the relative risk increases. Nevertheless, the relative risk of the integrated processes is below that of the non-integrated case for almost all the cases, except when both processes are fully integrated in a separated way.

Table 8
Results for the safety assessment in biodiesel process.

Scenario	Number of Heat Exchangers in the network	Number of hazardous streams in the process	Hazardous streams shared with bioethanol process	Relative risk
No integration	0	4	0	0.21
I	4	12	1	0.21
II	4	12	0	0.26
III	3	6	1	0.21
IV	3	6	2	0.19
V	5	6	4	0.18

Table 9
Results for the safety assessment in bioethanol process.

Scenario	Number of Heat Exchangers in the network	Number of hazardous streams in the process	Hazardous streams share with biodiesel process	Relative risk
No integration	0	7	0	0.19
I	4	10	1	0.18
II	3	9	0	0.24
III	2	6	1	0.12
IV	4	7	2	0.104
V	4	10	4	0.163

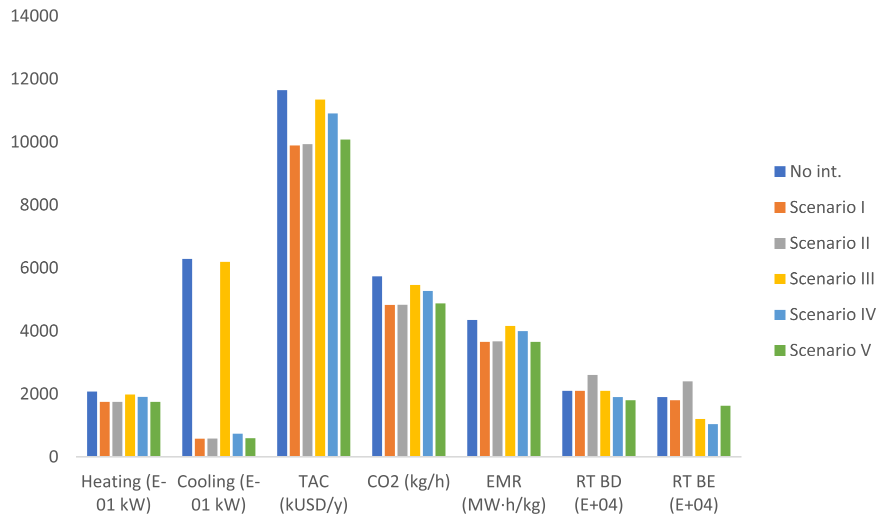

Fig. 9. Results summary.

5. Conclusions

Energy integration has a positive effect on utilities' requirements, reducing both heating and cooling needs. Commonly, this increases the capital costs, although for the analyzed processes this effect is not observed, due to changes on the kind of material required for the heat exchangers. Additionally, the integration reduces the emissions of carbon dioxide associated with the process, enhancing the energy use in the process. As the number of integrated streams increases, these effects are more evident. In the case of the safety assessment, it has been observed that the lowest relative risk is obtained for an intermediate number of integrated streams. Thus, a compromise can be found for all the indicators when partial integration occurs. This is the first time on which the HPSI index is used to assess safety aspects on integrated process. According to the assessment, the safety of the integrated processes could be enhanced if the less dangerous streams are preferred when proposing integration arrangements. Including such constraint may allow increasing the integration without worsening the safety of the process. Nevertheless, the complexity of the integrated network may have a negative impact on the flexibility and dynamic behavior of the process, thus, further studies related to the dynamic performance of the integrated processes are required.

CRediT authorship contribution statement

Juan Gabriel Segovia-Hernánde: Conceptualization, Methodology, Validation, Formal Analysis, Investigation, Writing – original draft, Writing – review & editing. **Fernando Israel Gómez-Castro:** Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Writing – review & editing. **Araceli Guadalupe Romero-Izquierdo:** Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Writing – review & editing. **Carolina Conde-Mejía:** Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Writing – review & editing. **Antioco López-Molina:** Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Writing – review & editing.

Declaration of Competing Interest

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

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