

Technical and Environmental Evaluation of an Integrated Biorefinery from Residual Biomass of Oil Palm Fruit to obtain Biobutanol and Biohydrogen

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Abstract: African palm fruit (*Elaeis guineensis*) has become one of the most widely studied and implemented raw material for vegetable oil production which is mainly used as food source and as a feedstock for biodiesel, production, however, unidirectional behavior of production chain causes waste accumulation and economic losses. In this research is presented an integrated biorefinery scheme based on oil palm fruit as raw material for the biobutanol and biohydrogen production, through simulation of each stage of the process using the commercial software UNISIM design. Technical analysis and environmental assessment using WAR algorithm were performed for five scenarios with different combinations of Oil Palm Empty Fruit Bunches (OPEFB) and Palm Pressed Fiber (PPF) in order to ensure the feasibility of its implementation at industrial scale as an alternative to expand the production chain. The best scenario resulting from this study was the mixture of OPEFB and PPF in a ratio 50:50 with a production rate of 6.900 kg/h of biobutanol, 680 kg/h of biohydrogen and 3.000 kg/h of other high value products. An environmental sustainability index (PEI) of 3.06/kg of product for the best scenario was obtained. Finally by energy integration, the potential for heating and cooling services saving was 9.5 and 92.2%, respectively. Environmental impact was reduced after the energy integration which shows that the integral use of the palm fruit through conversion processes in an integrated biorefinery is a technical and environmentally viable alternative to obtain biobutanol and biohydrogen as main products.

Key words: Biorefinery, palm pressed fiber, oil palm empty fruit bunches, heat integration, WAR algorithm, biorefinery

INTRODUCTION

The search for an integral use of biomass increases the alternatives of process improvement and optimization for the development of new bioindustries and processing technologies. Palm fruit has become one of the most widely studied and implemented raw material for obtaining oil which are mainly destined to the biodiesel production (Udonne *et al.*, 2016), however, linear behavior of production chain causes waste accumulation and environmental impacts when it is released without proper treatment as well as economic losses (Umor *et al.*, 2016). In this sense, the lignocellulosic material of the palm fruit is divided into two major types, oil palm empty fruit bunch and palm pressed fiber which have different compositions of hemicellulose, cellulose and lignin. Chemicals obtained from cellulose and hemicellulose includes alcohols, hydrogen, additives, acetone and weak acids by various fermentation processes that have been studied in the field of biotechnology in order to make it easier and practical

on an industrial scale as a sustainable alternative. One of the products that attract attention in recent years is biobutanol since its properties give it interesting characteristics such as lower volatility and explosiveness as well as a higher flash point and energy content. It is less corrosive and less hygroscopic and also can be easily mixed with gasoline and diesel (Michael *et al.*, 2011). On the other hand, biohydrogen has also been established as a viable alternative to fossil fuels due to its combustion, in addition only produces water as waste that can be used as a raw material for the production of other compounds, such as propanediol (Azman *et al.*, 2016). ABE fermentation has been one of the most studied processes for the bioethanol production due to high yields reached (Zanirun *et al.*, 2015; Du *et al.*, 2016). Ibrahim *et al.* (2015) focused on the use of enzymatic saccharification and simultaneous ABE fermentation using empty fruit bunches and pretreated fiber in order to produce biobutanol and biohydrogen. This study proposes to analyze the Potential of Palm Oil Fruit

Biomass (OPEFB and PPF) combinations under the concept of biorefinery to obtain biobutanol, biohydrogen and other added value products for different scenarios, evaluating its environmental and technical performance to make energy integration.

MATERIALS AND METHODS

Raw material: Palm Oil Empty Fruit Bunches (OPEFB) and Palm Pressed Fiber (PPF) are the main components of the residual biomass obtained in the oil palm industry by Ameera *et al.* (2016) and Faisal *et al.* (2016), the residual biomass stream was simulated based on the dry composition of the lignocellulosic material as it is shown in Table 1.

Simulation and process description: The simulation was performed using the software UNISIM design. The basis of calculation was taken as 5.000 t/day of clusters of fresh palm fruit, 23.8% correspond to OPEFB and PPF (Contreras *et al.*, 2015). Lignocellulosic components (cellulose, hemicellulose and lignin) as well as other compounds such as glucose and xylose were simulated from the properties reported by the National Renewable Energy Laboratory US. Thermodynamic fluid package NRTL (Non-Random Two Liquid) was chosen because of it gives very good predictions and it is suitable for processes with lignocellulosic material (Wooley and Putsche, 1996).

Biobutanol production: Lignocellulosic material was converted into biobutanol, biohydrogen and other byproducts via enzymatic saccharification with cellulase and ABE fermentation with the microorganism *C. acetobutylicum* simultaneously (Ibrahim *et al.*, 2015). Process presents three major steps: chemical pretreatment of biomass using sodium hydroxide, enzymatic hydrolysis and anaerobic fermentation. According to Jong and Gosselink (2014), alkaline pretreatment causes a high increase in surface area, hemicellulose and lignin removal and low inhibitor formation. The pretreatment step was simulated according to the conditions presented by Jaisamut *et al.* (2013). Conversion rates of cellulose, hemicellulose and lignin were set at 11.55, 32.37 and 54.22%, respectively and according to the solid losses reported by Dong (2014) for alkaline pretreatment of corn fiber, whose composition of lignocellulosic material is similar to oil palm biomass. Prior to enzymatic saccharification process and ABE fermentation, a stage of neutralization reaction was conducted for the pretreatment effluent with H_2SO_4 in order to ensure the best conditions for hydrolysis (Ashila, 2012).

Table 1: Chemical composition of residual biomass (Wooley and Putsche, 1996)

Biomass	Cellulose (%)	Hemicellulose (%)	Lignin (%)
OPEFB	30.4	21.7	18.1
PPF	33.9	26.1	27.7

Table 2: Reactions involved in the production of biohydrogen (Makinen *et al.*, 2012)

Stages	Reactions
Pretreatment and acid hydrolysis	Cellulose+ H_2O →Glucose Hemicellulose+ H_2O →Xylose
Fermentation	3Xylose+5 H_2O →5 Acetic acid 10 CO_2 +10 H_2 6Xylose→5Butyric acid 10 CO_2 +10 H_2 Glucose+2 H_2O →2 Acetic acid+2 CO_2 +4 H_2 Glucose→Butyric acid+2 CO_2 +2 H_2

Simultaneous Saccharification and Fermentation (SSF) was performed in a biological conversion reactor at 37°C and 1 atm. The conversion rate of enzymatic hydrolysis reactions by commercial cellulase action (Celluclast 1.5 L, ratio: 0.006 kg cellulase/kg solids) was set at 93.1% of cellulose (Jaisamut *et al.*, 2013) and 60% hemicellulose (Sanchez and Cardona, 2008). Finally, the conversion of fermentable sugars by the microorganism *C. acetobutylicum* (0.15 kg/kg solids) were stipulated in 57.81, 27.36 and 7.35% for biobutanol, acetone and ethanol, respectively, according to Naleli (2016). Similarly, the formation of undesired products in ABE fermentation such as the butyric acid and acetic acid in small quantities were considered.

Biohydrogen production: Biohydrogen production was simulated from 5-carbon sugars (xylose). For this case, an acid pretreatment for biomass with H_2SO_4 dilute at 1.5%, 160°C and 1 MPa with solids loading in the reactor of 10% (g dry substrate/g mixture) was performed (Sanchez and Cardona, 2008). The fermentation was conducted in a biological conversion reactor at 40°C and 1 MPa in the presence of *C. beijerinckii* YA001 (0.15 kg/kg solids) with a mole ratio of 2.31 H_2 /xylose. Also, the glucose mole ratio for the hydrogen production was set at 1.7 H_2 /glucose (Makinen *et al.* 2012) (Table 2).

Technical analysis: Five scenarios with different combinations of OPEFB and PPF for the Bio Butanol (BB) production were considered and the rest of raw material was used for the Bio Hydrogen (BH) production. The main objective was to maximize the amount of desirable product based on the OPEFB and PPF compositions of cellulose and hemicellulose, the amount of fermentable sugars with 6 or 5 carbons used to obtain biobutanol and biohydrogen, respectively was selected based on this aim. The scenarios are as follows:

- Scenario 1: BB from OPEFB
- Scenario 2: BB from PPF

- Scenario 3: BB from 20% OPEFB-80% PPF
- Scenario 4: BB from 50% OPEFB-50% PPF
- Scenario 5: BB from 80% OPEFB-20% PPF

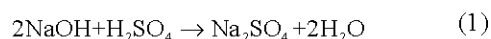
Environmental assessment: The environmental analysis was performed for each case and scenario by estimating the Potential Environmental Impact (PEI) through the WAR algorithm, developed by the Environmental Protection Agency of the United States. The algorithm was used to calculate the index of overall environmental impact that is mainly based on the harmfulness of the substances involved in the process if they are released into the environment. The PEI is evaluated under 8 impact categories: Human Toxicity Potential by Ingestion (HTPI), Human Toxicity Potential by Dermal Exposure and inhalation (HTPE), Terrestrial Toxicity Potential (TTP), Aquatic Toxicity Potential (ATP), Global Warming Potential (GWP), Ozone Depletion Potential (ODP), Photochemical Oxidation Potential (PCOP) and Acidification Potential (AP) (Gonzalez-Delgado and Peralta-Ruiz, 2016) which are calculated based on the inlet and output mass and energy flows of the process. The heating service is assumed as natural gas due to is cheaper to generate electricity and cleaner compared to coal and oil.

Heat integration: Heat integration was performed in the scenario that presented a balance between high productivity and low environmental impact, using the pinch analysis methodology. In the first instance, the Cold streams (Ci) and Hot streams (Hi) with their respective mass flows, heat capacities and changes in temperature were identified and set a minimum delta T = 10°C in order to reach a thermal equilibrium and heat transfer feasible. Subsequently, the algebraic method known as “cascade diagram” which is based on the solution algorithm developed by Linnhoff and Flower

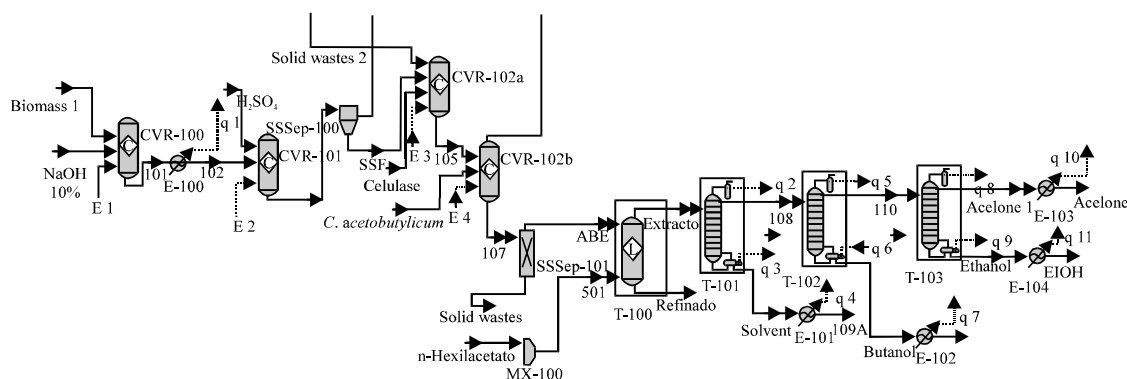
(1978) was implemented. Through the cascade diagram the minimum requirements of external Heating (QH_{min}) and external Cooling (QC_{min}) were obtained. Grand Composite Curve (GCC) was developed thus obtaining the temperature at which industrial services are required. With this, it was necessary to determine the type of industrial service. Then a heat exchange network that allows obtaining the greatest possible energy savings in the process according to the points made by El-Halwagi (2011) guaranteeing the minimum set temperature difference was presented. Finally, the scheme proposed for the biorefinery designed was modified by adjusting the heat exchange network obtained in order to integrate the energy process.

RESULTS AND DISCUSSION

Process simulation: Figure 1 shows the pretreatment step simulation in a Conversion Reactor (CVR-100) where total solids loss was 29% based on the weight of the initial dry biomass. Because of the hydrolysis step requires optimum conditions, the reactor effluent stream was neutralized (CVR-101) by Eq. 1, so that, the sodium hydroxide was consumed with H_2SO_4 at 98% to produce soluble Na_2SO_4 . The pretreated and neutralized biomass passed to a Solid-liquid Separator (SSSep-100) in order to extract the hydrolyzed xylose in the pretreatment and traces of soluble glucose and lignin (stream 104) from the solids (SSF stream) where the solids go out with a minimum percentage of moisture and the liquid stream without suspended solids:



For practical purposes, simultaneous enzymatic saccharification and fermentation were simulated in two continuous conversion reactors (CVR-102 a and b) at



Variables	LL (T-100)	D1 (T-101)	D2 (T-102)
No. of stages	15	25.0	22
Inlet stage	1	10.0	13
Reflux ratio	-	2.2	2

In order to separate the organic solvents obtained from the fermentation in stream 107 (butanol, acetone and ethanol), conventional separation and purification technology based on Liquid-Liquid (LL) extraction followed by a Double Distillation (D1-D2) were implemented. The operating conditions at 1 atm pressure are shown in Table 3 where each tower was designed in order to meet the requirements of the process.

Properties	1-Butanol	Bu-OH
Composition (W/W)	99.4%	99.6%
Boiling point (°C)	116-118	117.9
Vapor pressure (kPa) @ 20°C	0.67	0.64
Density (g/cm ³) @ 20°C	0.809-0.813	0.815
Molar mass (g/mol)	74.12	74.02

Biohydrogen production process shown in Fig. 2 uses the stream (104) of fermentable 5-carbon (xylose) sugars and traces of effluent from the Solid-liquid Separator (SSSep-100). Palm pressed fiber was used as a raw material in this process. Hydrogen production presents the same stages of biobutanol production: pretreatment, fermentation and separation.

536

Table 5: Comparison of amount of product, energy and waste obtained in each scenario

Variables/Units	Scenario 1	Scenario 2	Scenario 3	Scenario 4	Scenario 5
Biobutanol (kg/h)	6,906.86	6,806.27	6,869.45	6,964.06	6,907.46
Bioethanol (kg/h)	1,361.80	1,346.15	1,357.24	1,369.65	1,363.99
Bioacetone (kg/h)	1,652.28	1,632.53	1,643.60	1,664.54	1,650.29
Biohydrogen (kg/h)	682.56	678.49	681.57	686.19	682.84
Energy (MJ)	296,214.95	311,028.57	303,917.23	299,228.57	308,182.42
Solids (kg/h)	18,132.00	17,518.42	16,991.95	17,848.78	17,307.66

at 1 MPa. Acid pretreatment of the biomass in the CVR-200 reactor presented 31% of solids loss percentage based on the weight of initial dry biomass. Previous neutralization of the effluent carried out in the CVR-201 reactor was also necessary for the fermentation step by the reaction described in Eq. 1. After the pretreated biomass was neutralized, a Solid-liquid Separator (SSSep-102) was used to remove the hydrolyzed xylose in the pretreatment and glucose traces of the nonreactive biomass (residual solids 2). In order to remove the Na_2SO_4 , both the liquid stream from the separator (208) and the stream 104 passed through a simulated filter as a split unit (X-100), obtaining an aqueous solution with 3.2% xylose. Fermentation was carried out at 40°C and 1 atm pressure. At the reactor top (CVR-202) a gas stream (210) of 8.116 kg/h was obtained, composed of 87.8% CO_2 , 6.5% H_2O , 5.4% H_2 and traces of glucose, acetic acid and butyric acid. A liquid stream (211) of 96.3% water was obtained from the bottom of the reactor with traces of glucose, soluble lignin, acetic acid and butyric acid. Subsequently, the biohydrogen separation step was carried out, the gas stream (106) from the fermenter in the ABE production also entered into the hydrogen recovery equipment (X-101) which is based on a Pressure differential Absorber (PSA) operating at 23.08 bars and 37.78°C obtaining hydrogen with a purity of 99.9%.

Technical analysis: Table 5 shows the amount of products obtained under the 5 scenarios. The difference is not pronounced however in scenario 4 was produced the highest amount of biobutanol and biohydrogen.

Environmental assessment: Figure 3 shows the results for environmental analysis for all 5 scenarios studied. The WAR algorithm works with two indexes to evaluate the PEI of the process: the PEI output (I_{out}) and generated (I_{gen}). Each index can be expressed as a function of time and mass of product, however, it is observed that both had the same trend due to the distribution of the components fractions (lignin, cellulose and hemicellulose) for the two types of biomass.

As it is observed having different mixtures of biomass lead to a small variation in the environmental impact indexes. In addition, scenario 4 presents the lower rates with a more negative generation of PEI, this is

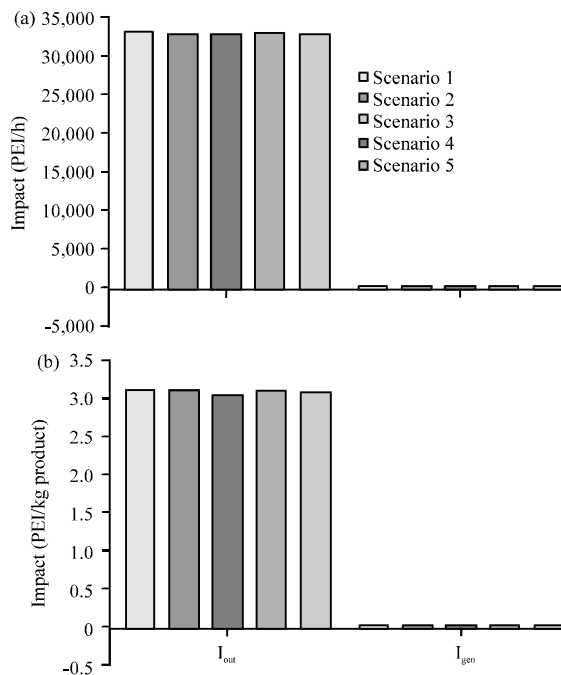


Fig. 3: Total PEI indexes per: a) Time and b) Mass of product under the 5 scenarios studied

because the potential impact of the substances that enter to the process are transformed into others that are not considered as waste and are non-fossil energy sources. Figure 4 are shows the different categories that affect the overall process PEI per mass of product. The most significant impacts were obtained under HTPI and TTP categories which are mainly represented by the toxicological properties of wastes and products generated. Solid wastes are rich in lignin (presents medium toxicity being an organic residue) with small traces of chemicals synthesized in the process, these two factors make a contribution to the considerable increase in these indicators. The PCOP category is directly affected by the gas rich in CO_2 that is vented to the atmosphere, so the index would decrease significantly if CO_2 were considered as a product and not as a residue.

Heat integration: According to the technical and environmental analysis, the ideal scenario is the fourth where the combination of biomass in 50:50 ratio is used.

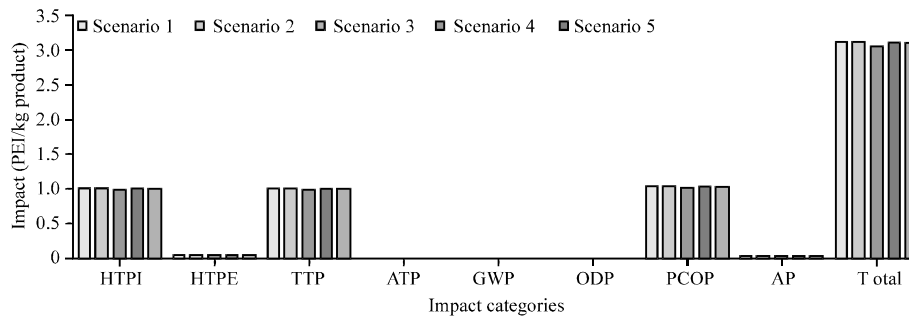


Fig. 4: Potential environmental impact leaving the system per mass of product under impact categories studied

Table 6: Streams of the biomass utilization process chosen for pinch analysis.

Streams	Mass flow (kg/sec)	Heat capacity (kJ/ kg°C)	FCP (kW/°C)	Supply temperature (°C)	Target temperature (°C)	Enthalpy (kW)
C ₁	1.9	4.13	8.0	30.00	80.00	398.6
C ₂	105.0	4.18	438.8	30.00	100.00	30,568.3
H ₁	8.5	2.40	20.4	170.80	30.00	2,507.0
H ₂	1.9	3.11	6.0	117.90	30.00	494.0
H ₃	0.5	2.15	1.0	55.80	30.00	30.7
H ₄	0.4	3.65	1.4	80.00	30.00	67.7

Table 7: Potential of energy integration of the process under scenario 4

Utilities	Base case (kW)	Integrated process (kW)	Potential savings (%)
Heating	30,966.9	27,915.8	9.9
Cooling	3,099.4	287.5	90.7

Table 6 shows the Cold (Ci) and Hot (Hi) streams selected from the simulation and the extracted data such as supply and target temperatures, mass flows and heat capacity of each stream. It should be noted that the selected ones are those that are not solid, do not present phase change and those that do not require too much energy to reach the desired temperature.

Due to the methodology used, a cascade diagram was built, from which a potential of energy integration was obtained for the heating and cooling utilities of the whole process of 9.9 and 90.7%, respectively as it is observed in Table 7. The saving potential of the heating utility is low because the Cold stream (C₂) has a very high enthalpy value in comparison with the other streams which means that C₂ has the capacity to supply the largest part of the cooling service above the pinch point and still cannot get enough heat, requiring 27,535.5 kW from an additional heating service. It is observed that cold streams are capable of cooling all hot streams but 287.5 kW of cooling utility is required because of no cold stream is available below the pinch point.

In order to know what types of industrial services meet the requirements of heating and external cooling, the large Composite Curve (GCC) observed in Fig. 5 was constructed. Due to the temperature range below the pinch is 35-25°C, it is possible to use cooling water as an external service to supply the required 287.5 kW. Above

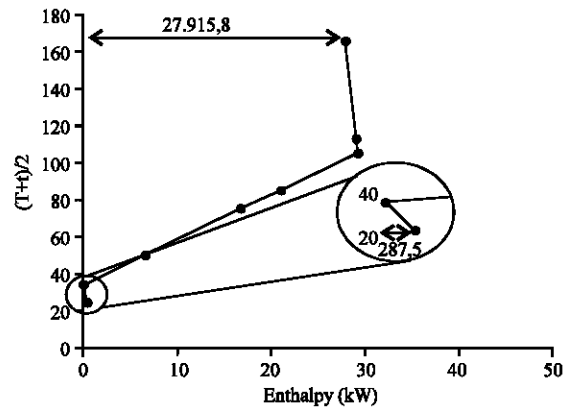


Fig. 5: Grand Composite Curve (GCC) of the process under scenario 4

the pinch, the temperature range is 50-170°C, so that, a medium pressure steam can be used as an external heating medium at 20 bar with a condensing temperature of 212°C. The heat exchange network designed and proposed for the energy integration of the biomass utilization process is shown in Fig. 6, based on the minimum temperature change stipulated (10°C). It is noted that above the pinch point all the hot streams are energetically integrated however, heaters in the C₁ and C₂ streams were required to reach the target temperature. In order to follow the population rule of streams above the pinch, the C₂ stream (the one with higher enthalpy) was divided into 3, each with the same transferable charge of energy. Also, it is observed that below this point there are no cold streams

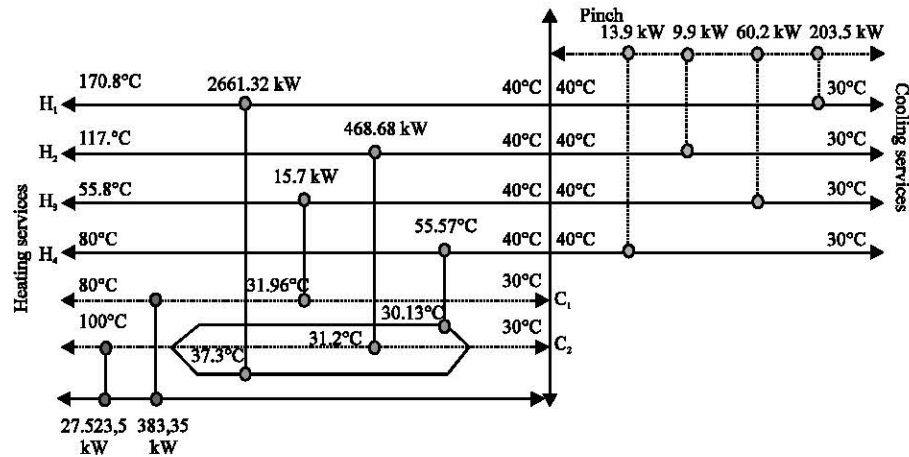


Fig. 6: Heat exchange network proposed for the process under scenario 4

Table 8: Potential of energy integration obtained in the simulation of the integrated process

Utilities	Integrated process (kW)	Saving potential (%)
Heating	28,035.7	9.5
Cooling	241.8	92.2

Table 9: Environmental indicators for Non-Integrated (N-I) and Integrated (I) process under scenario 4

PEI/kg	HTPI	HTPE	TTP	ATP	GWP	ODP	PCOP	AP	Total
N-I	0.984	0.035	0.984	0.00274	0.00325	0	1.03	0.0282	3.06
I	0.984	0.035	0.984	0.00246	0.00162	0	1.03	0.0114	3.04

that can be integrated with the hot streams, therefore, it became necessary to set heaters to each hot stream in order to achieve the target temperature of 30°C.

Finally, the implementation of the heat exchange network in reality does not necessarily reach the savings percentages determined by the pinch analysis even when the process is optimized, just as it happens when the process is simulated with the proposed exchange network. As it is shown in Table 8, the percentages of savings in industrial services are similar to those obtained by the pinch analysis (Table 7). For the simulation of the integrated process were used tube and shell heat exchangers available in the software and the cooling and heating services were simulated with cooler and heater, respectively. The total number of heat exchangers increased from 10-14.

In addition, Table 9 shows a comparison of environmental assessment for the non-integrated and integrated process where it is observed a decrease under ATP, GWP and AP categories which are related to atmospheric impacts.

CONCLUSION

This study presents the simulation of a biorefinery for the biobutanol and biohydrogen production as main products and bioacetone/bioethanol as high value by-products, from Oil Palm Empty Fruit Bunches (OPEFB) and Palm Pressed Fiber (PPF). A technical analysis under

5 scenarios with different combinations of biomass was performed, environmental assessment using the WAR algorithm and energy integration for the scenario with the best results were also performed. The proposed topology of biorefinery was modified based on the heat exchanger network obtained in order to compare the saving potential of energy and environmental impacts. For the scenarios evaluated, the most feasible case was found for a biomass ratio of 50:50 where 6,964.06 kg/h of biobutanol was obtained. In addition, the total PEI was 3.06 PEI/kg of product and the highest impact values were obtained for HTPI and TTP categories due to solid wastes are rich in lignin with small traces of chemicals synthesized in the process. On the other hand, heat integration potential for this scenario in terms of heating and cooling utilities was 92.2 and 9.5%, respectively. Results showed that heat integration reduces significantly the industrial services and generate less environmental impact. Therefore, obtaining biobutanol and biohydrogen in an oil palm fruit based-biorefinery is a favorable alternative to implement under operating conditions and process steps presented for the case selected as suitable due to high productivity, low environmental impact and energy savings.

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