Technological, economic and environmental evaluation of rice husk gasification in a biorefinery context to produce indirect energy as jet fuel

Evaluación tecnológica, económica y medioambiental de la gasificación de la cascarilla de arroz en un contexto de biorefinería para producir energía indirecta como combustible de jet

Avaliação tecnológica, econômica e ambiental da gaseificação da casca de arroz em um contexto de biorrefinagem para produzir energia indireta como combustível para jatos

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Abstract

Higher alcohol 1-octanol was evaluated as jet fuel potential. The synthesis of the 1-octanol was modeled and the technological, economic and environmental evaluation of the global production process of the rice husk gasification was performed. The best operating conditions to 1-octanol synthesis were obtained in packed bed reactor PBR using Matlab software. Mass and energy balances were calculated using Aspen Plus Software. Economic assessment was developed using Aspen Process Economic Analyzer Software. Environmental impact evaluation was carried out using the waste reduction algorithm WAR. Process yield was 0.83 kg of 1-Octanol by kg of rice husk. Total production cost obtained was USD 0.957 per kg of 1-octanol and the total PEI of product leave the system is 0.08142 PEI/kg with a PEI mitigated of 12.97 PEI/kg. Production process of high alcohols from rice husk shows a high potential technological, economical and environmental as a sustainable industry at take advantage of an agroindustrial residue and transformed in products with added value and energy. 1-octanol as jet fuel has a potential but need to be more studied for direct use in jet motors.

Key-words: higher alcohols, 1-octanol, jet fuel, technological evaluation, economic evaluation, environmental evaluation.

Resumen

El presente artículo evaluó el potencial del 1-octanol como combustible jet. Se modeló la síntesis del 1-octanol y se realizó la evaluación tecnológica, económica y ambiental del proceso de producción de 1-octanol a partir de la gasificación de la cascarilla de arroz. Las mejores condiciones de funcionamiento para la síntesis de 1-octanol, se obtuvieron en un reactor de lecho empacado (PBR) utilizando el software Matlab. Se calcularon los balances de masa y energía empleando el software Aspen Plus. La evaluación económica se desarrolló utilizando Aspen Process Economic Analyzer Software. La evaluación del impacto ambiental se llevó a cabo utilizando el algoritmo de reducción de residuos WAR. El rendimiento del proceso fue de 0,83 kg de 1-octanol por kg de cascarilla de arroz. El costo total de producción obtenido fue de USD 0.957 por kg de 1-octanol y el PEI total del producto es de 0.08142 PEI / kg con un PEI atenuado de 12.97 PEI / kg. El proceso de producción de alcoholes altos a partir de cascarilla de arroz muestra un alto potencial tecnológico, económico y ambiental, como una industria sostenible en aprovechar un residuo agroindustrial y transformarlo en productos con valor agregado y energía. El 1-octanol tiene potencial como combustible jet, pero necesita ser más estudiado para su uso directo en motores de avión.

Palabras clave: alcoholes altos, 1-octanol, combustible de jet, evaluación tecnológica, evaluación económica, evaluación ambiental.

Resumo

O maior álcool 1-octanol foi classificado como potencial de combustível de jato. A síntese de 1-octanol foi modelada e a avaliação tecnológica, econômica e ambiental do processo de produção global da gaseificação do escudo de arroz foi realizada. As melhores condições operacionais para a síntese de 1-octanol foram obtidas no reator embalado no leito, usando o software MATLAB. Os balanços de massa e energia foram calculados usando o software Aspen Plus. A avaliação econômica foi desenvolvida usando o software Aspen Economic Process Analyzer. A avaliação do impacto ambiental foi realizada utilizando o algoritmo de redução de guerra. O rendimento do processo foi de 0,83 kg de 1-octanol por kg de casca de arroz. O custo total da produção foi de USD 0,957 por kg de 1 octanol e o PEI total do sistema de licença do produto é de 0,08142 IEP / kg com IEP mitigado de 12,97 IEP / kg. O processo de produção de álcoois de casca de arroz mostra um alto potencial tecnológico, econômico e ambiental como uma indústria sustentável que aproveita um resíduo agroindustrial e é transformado em produtos com valor agregado e energia. O 1-octanol como combustível para jatos tem potencial mas precisa ser estudado para uso direto em motores a jato.

Palavras chave: álcoois superiores, 1 octanol, combustível para jatos, avaliação tecnológica, avaliação econômica, avaliação ambiental.

Introduction

The jet fuel demand in the United States is about three billion gallons annually that represent 10% of the U.S market for aviation fuel (Balster *et al*., 2008). The characteristics of jet fuel must do a dual role of fuel and coolant over ranges of combustion chambers in terms of temperature and pressure. The recent research is focused on the production hydrocarbons with jet fuel characteristics (Kumar & Sung, 2010). The higher alcohols synthesis is a basic point in the C_1 chemistry. The works in this area are focused on the production of octane booster for cleaner jet fuels and gasoline (Herman, 2000).

From the thermodynamic point of view, the reactions of high alcohols formation are profitable to lower temperatures and higher pressures because these reactions are exothermic. From the kinetic point of view, temperatures about 600 ^oK improves the reactions (Herman, 2000). The reactions require a special catalyst to determine operation conditions containing alkali species (Breman, Beenackers, & Oesterholt, 1994; Calverley, 1989; Kulawska & Skrzypek, 2001; Surisetty, Dalai, & Kozinski, 2010; Surisetty, Dalai, & Kozinski, 2011). The reaction mechanisms depending on the process conditions and catalyst used, alcohols are synthesized using iso-synthesis, variants of Fisher–Tropsch synthesis, oxo-synthesis involving the hydroformylation of olefins, and homologation of methanol and low molecular weight alcohols to make higher alcohols (Surisetty, Dalai, & Kozinski, 2010). The following are the chemistry reactions involved in the higher alcohols synthesis from syngas:

$$CO + 2H_2 \longleftrightarrow CH_3OH \quad (1)$$
$$CO + H_2O \longleftrightarrow CO_2 + H_2 \quad (2)$$
$$CH_3OH + CO + 2H_2 \longleftrightarrow CH_3CH_2OH + H_2O \quad (3)$$
$$C_nH_{2n+1}OH + CO + 2H_2 \longleftrightarrow CH_3\left(CH_2\right)_n OH + H_2O \quad (4)$$
$$2CH_3OH \longleftrightarrow CH_3CH_2OH + H_2O \quad (5)$$
$$2CH_3OH \longleftrightarrow \left(CH_3\right)_2 CO + H_2O \quad (6)$$
$$CH_3OH + CO \longleftrightarrow CH_3CHO + H_2O \quad (7)$$
$$\left(CH_3\right)_2 CO + H_2 \longleftrightarrow \left(CH_3\right)_2 CHOH + H_2O \quad (8)$$
$$2CH_3CHO \longleftrightarrow CH_3COOCH_2CH_3 \quad (9)$$

The reactions schemes for higher alcohols synthesis from methanol are:

$$CH_3OH + s \rightarrow CH_3OH_s \quad (10)$$
$$CH_3OH_s + s \rightarrow CH_3O_s + H_s \quad (11)$$
$$CH_3O_s + s \rightarrow CH_3CH_2O_s + O_s \quad (12)$$
$$CH_3O_s + CH_2O_s \rightarrow CH_3CH_2O_s + O_s \quad (13)$$
$$CH_3CH_2O_s + Hs \rightarrow CH_3CH_2OH_s + s \quad (14)$$
$$CH_3CH_2OH_s \rightarrow CH_3CH_2OH + s \quad (15)$$
$$\mathrm{CO}_s + O_s \rightarrow CO_{2,s} + s \quad (16)$$
$$\mathrm{CO}_{2,s} \rightarrow CO_2 + s \quad (17)$$
$$CO + s \rightarrow CO_s \quad (18)$$
$$H_2 + s \rightarrow H_{2,s} \quad (19)$$

Alcohols can be used as an alternative jet or components fuels for many reasons, such as:

- Reduction of fuel cost
- Reduction of toxic exhaust emissions
- To enhanced of overall energy efficiency
- Reduction of greenhouse gas emissions
- Social reasons

In this paper, the higher alcohol 1-octanol is evaluated as jet fuel potential. 1-octanol synthesis is modeled and the technological, economic and environmental evaluation of the production process is performed from rice husk gasification.

Synthesis of the 1-octanol

The high alcohols production was modeling according to the kinetic expression proposed by Calverley,1989. This kinetic model assumes the production of higher alcohols from CO, H2 and methanol promoted by copper/zinc-oxide/chromia catalysts. These catalysts suggest to inhibition for CO_2 in the gas are because a catalyst poisoning. The global chemical reactions and the kinetic rates are shown below:

Global Reactions:

$$nCO + 2nH_2 \rightarrow C_nH_{2n+1}OH + (n-1)H_2O \quad (20)$$
$$(2n-1)CO + (n+1)H_2 \rightarrow C_nH_{2n+1}OH + (n-1)CO_2 \quad (21)$$
$$(n-1)CO + (n-1)H_2O \longleftrightarrow (n-1)CO_2 + (n-1)H_2 \quad (22)$$
$$nCH_3OH \rightarrow C_nH_{2n+1}OH + (n-1)H_2O \quad (23)$$

Kinetic Expression:

$$r_{HA} = \left[\frac{P_{CO} P_{H_2}^{\frac{1}{2}}}{A + BP_M + F\left(\frac{P_{CO_2}}{P_{CO}}\right)} + CP_M \left(P_{H_2}\right)^{\frac{-3}{2}} + \frac{P_{CO_2} P_{H_2}^{\frac{1}{2}}}{D + E\left(\frac{P_{CO_2}}{P_{CO}}\right)} \right]^2 \quad (24)$$

Where A=4578, B=2794, C=0.338, D=35.62, E=8228, F=2.44x10^6. P_{CO}, P_{H2}, P_M, P_{CO2} are the partial pressure of CO, H_2, methanol, CO_2 in atm respectively. The kinetic expression (r_{HA}) is in mmol h^{-1} kg^{-1}.

All reactions were performed in the gas state therefore a packed bed reactor PBR was modeled by the high alcohol 1-Octanol (n=8 in the reactions). The molar balances in the PBR are:

Molar Balances PBR:

$$\frac{dF_{CO}}{dW} = -\left(\frac{16}{3}\right) r_{HA} \quad (25)$$

$$\frac{dF_{H_2}}{dW} = -\left(\frac{32}{3}\right) r_{HA} \quad (26)$$

$$\frac{dF_{CH_3OH}}{dW} = -\left(\frac{8}{3}\right) r_{HA} \quad (27)$$

$$\frac{dF_{C_8H_{17}OH}}{dW} = r_{HA} \quad (28)$$

$$\frac{dF_{H_2O}}{dW} = \left(\frac{21}{3}\right) r_{HA} \quad (29)$$

F is the molar flow of CO, H_2, methanol (CH_3OH), 1-octanol ($C_8H_{17}OH$) and H_2O. W is the catalyst charge in kg.

Methodology

The 1-octanol synthesis was simulated and modeling in the software commercial Matlab R2008b using the toolbox ode23s and a sensitivity analysis to solve the molar balances and the best operating conditions of the reaction zone PBR. These operation conditions were: the yield of 1-octanol to CO, the reaction pressure, the CO_2/CO feed ratio, the H_2/CO feed ratio, the CH_3OH feed ratio.

Aspen Plus software (AspenTech: Cambridge, MA) was used to simulate the 1-octanol production global process. The physicochemical properties were obtained from the National Institute of Standards of Technology NIST (NIST, 2005) and the group-contribution method developed by Gani *et al.* (Marrero, 2001) at three different levels. The on random two-liquid NRTL thermodynamic model was utilized to calculate the activity coefficients in the liquid phase, and the Hayden–O'Connell equation of state was used to model the vapor phase. Mass and energy balances were calculated by simulation.

Economic evaluations were performed using Aspen Process Economic Analyzer in the Colombian context -with an annual interest rate of 17% and a tax rate of 33%-. A straight-line depreciation method was used over a 12-year period of analysis. For feedstock prices, the international reports from ICIS pricing were employed; operating charges such as operator and supervisor labor costs were defined for Colombia at 2.14 and 4.29 USD/h, respectively. Electricity, potable water, low and high steam pressure costs were 0.1 USD/kWh, 1.252 USD/m^3, and 8.18 USD/ton, respectively.

The environmental impact was assessed with the waste reduction WAR algorithm -developed by the U.S. Environmental Protection Agency-, to estimate the potential environmental impact PEI generated in the process considering eight environmental impact categories: human toxicity potential by ingestion HTPI, human toxicity potential by dermal and inhalation exposure 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. The mass flow rate of each component in the process streams was multiplied by its chemical potential to determine its contribution to the potential environmental impact categories (Cabezas, Bare, & Mallick, 1999; Cardona, Marulanda & Young, 2004).

Results

The results of modeling and simulated the PBR to produce 1-octanol are shown in the Figures 1-5 in order to view the influence of reaction pressure, the CO_2/CO feed ratio, the H_2/CO feed ratio, the CH_3OH feed ratio on the yield of 1-octanol to CO and the CO, H_2 and methanol conversions. Table 1 summarizes the best operating conditions in the 1-octanol synthesis.

The global process to produce 1-octanol from rice husk is described in the Figure 6. Rice husk is fed to dryer tunnel with air to reduce the moisture content. The free water rice husk is crushed to reduce the particle diameter and ensure a superficial area suitable to gasification. The water in the air output is condensed to use in the global process as fluid service. The rice husk dust is fed to gasification unit to produce syngas -CO and H_2 mainly-, with which is generated high pressure vapor HPV in a heat exchanger and the rice husk ash is collected.

The exhausted syngas is the reactive in 1-octanol synthesis with methanol promoted copper/zinc-oxide/chromia catalysts. The reaction is performed in a PBR to produce 1-octanol and water mainly with a yield of 0.3 mole of 1-octanol per mole of CO.

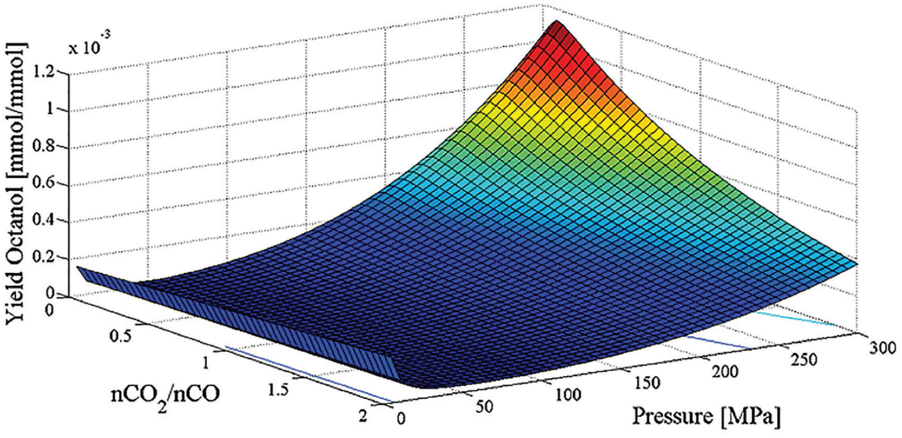

Figure 1. Influence of reaction pressure, the CO_2/CO feed ratio on the 1-Octanol Yield.

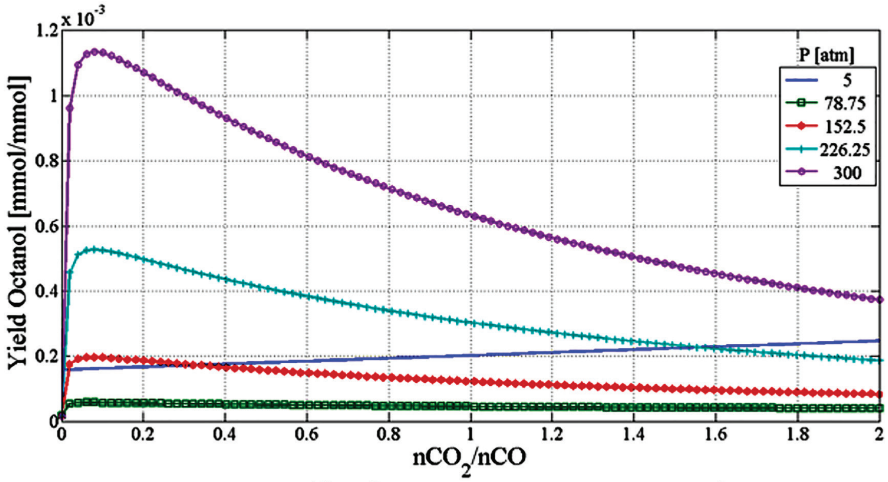

Figure 2. Influence of CO_2/CO feed ratio on the 1-Octanol Yield to different pressures.

The methanol excess is fed to other PBR to make the condensation methanol reaction to convert in 1-octanol. The unreacted gas and methanol traces are separated in an evaporator flash unit and these are sent to furnace to produce hot gas. The CO_2 and H_2O vapor bulk is fed to heat exchanger to produce other streams with HPV which is mixed and sent to a turbine to generate electricity. The 1-octanol-water bulk is separated in extraction and distillation stages to take advantage of liquid-liquid vapor equilibrium of the system water-methanol-1-octanol.

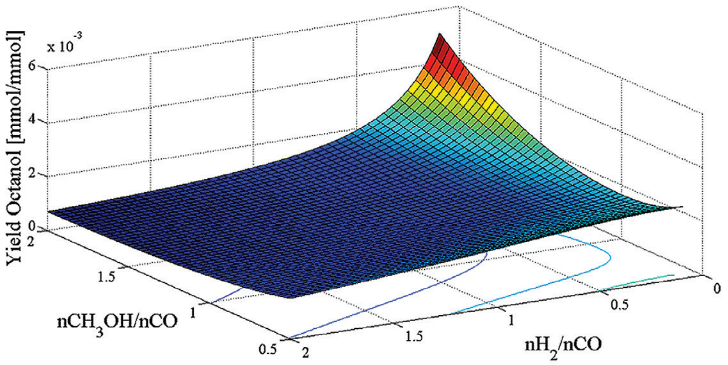

Figure 3. Influence of H_2/CO and CH_3OH/CO feed ratio on the 1-Octanol Yield.

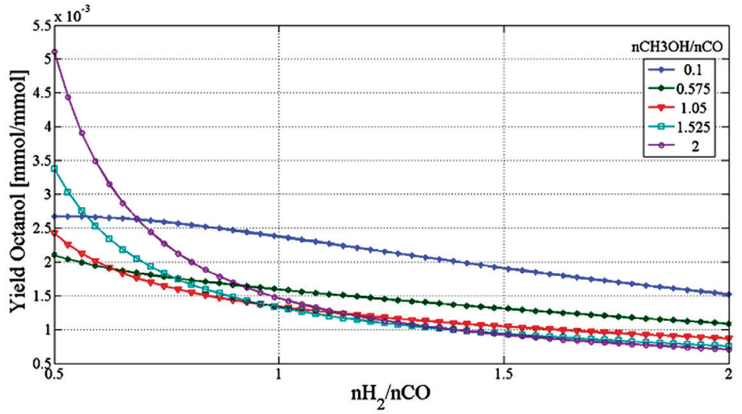

Figure 4. Influence of H_2/CO on the 1-Octanol Yield to differences CH_3OH/CO feed ratios

Mass and energy balances of the process structure are shown in the Table 2. The result of the Process Yield to 1-Octanol is: 0.83 kg of 1-Octanol by kg of Rice husk and Process Yield to Rice husk Ash: 0.12 kg of Ash by kg of Rice husk.

Economic evaluation is shown in the Table 3. The total production cost obtained was USD 0.957 per kg of 1-octanol with a Total Capital Cost: USD 9'571,280. The distribution of the total production cost is observed in the Figure 7.

The environmental evaluation is illustrated in the Figure 8. The total PEI of product leave the system is 0.08142 PEI/kg with a PEI mitigated of 12.97 PEI/kg.

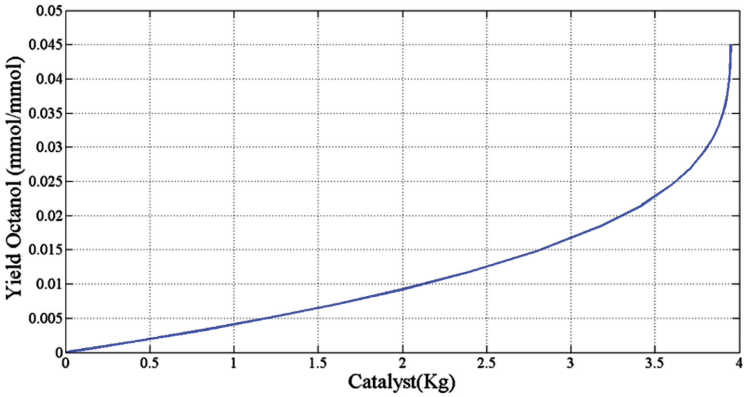

Figure 5. Profile 1-Octanol Yield vs Catalyst charge.

Table 1. *Best operation conditions found in the 1-octanol synthesis.*

Operation conditions	Value
Yield: 1-octanol to CO	0.043 (mol)
CO conversion	22.91%
H_2 conversion	85.92%
Methanol conversion	5.73%
CO_2/CO feed ratio	0.081 (mol)
H_2/CO feed ratio	0.50 (mol)
CH_3OH/CO feed ratio	2.00 (mol)
Pressure	300 atm

Table 2. *Mass and Energy Balances for 1-octanol production from rice husk.*

Feedstock	Flow (kg h^{-1})
Rice husk	914
Water	1549.3
Air	4000
Methanol	1550.3
Products	**Flow (kg h^{-1})**
1-Octanol @99.9%	757.9
Rice husk Ash	116.1
Waste water	2379.1
Exhausted Gas	4744.6
Energy	**Flow (kW)**
Energy Demand	386.75
Power Generated	227.2

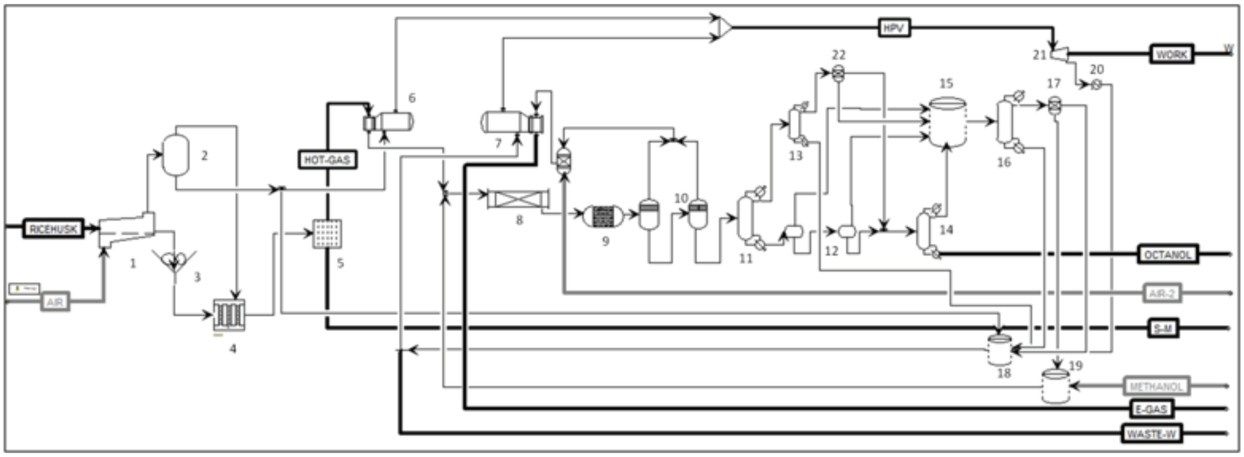

Figure 6. Process Diagram Flow of 1-octanol production Captions: 1: dryer, 2: dehumidifier, 3: crusher, 4: gasification unit, 5: gas-solid filter, 6 and 7: heat exchanger, 8: 1-octanol synthesis from syngas and methanol PBR, 9: 1-octanol synthesis from methanol only PBR, 10: flash units, 11: distillation tower, 12: extraction tanks, 13: distillation tower, 14: distillation tower, 15: methanol aqueous tank, 16: distillation tower, 17: absorber, 18: water tank, 19: methanol tank, 20: cooler, 21: turbine, 22: split.

Table 3. *Economic Evaluation for 1-octanol production from rice husk*

Cost	Value USD kg^{-1} of 1-Octanol produced
Raw Materials	0.745
Operating Labor and Maintenance	0.015
Utilities	0.056
Operating	0.026
Depreciation expenses	0.115
Total Production Cost	**0.957**

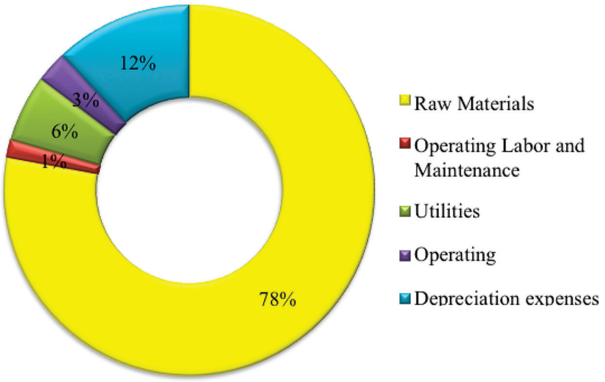

Figure 7. Production Cost Distribution by 1-octanol from rice husk

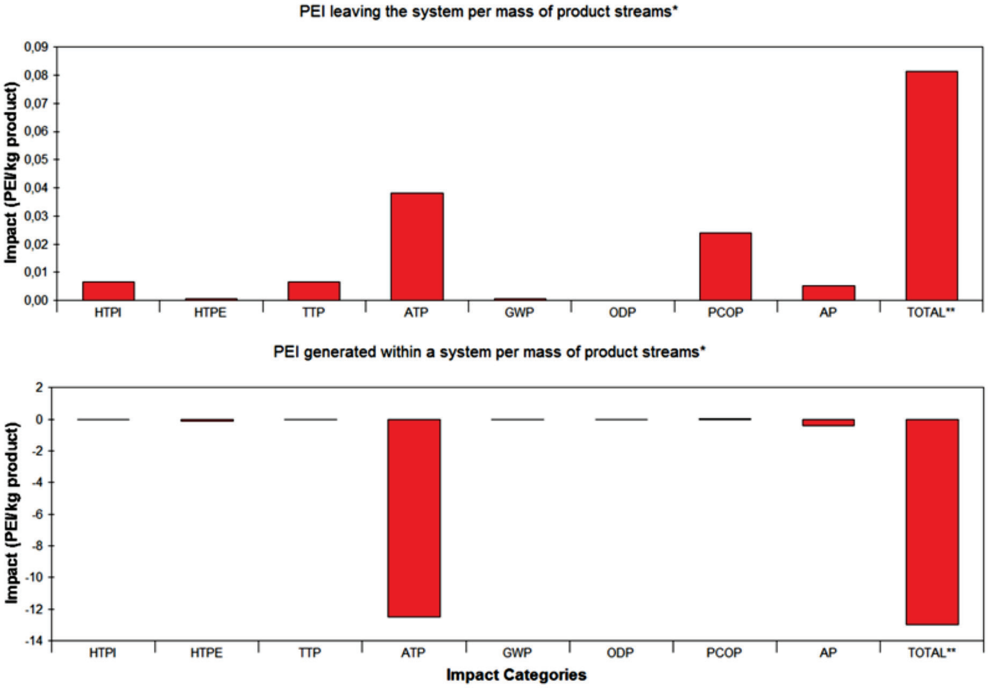

Figure 8. Environmental evaluation to 1-octanol production process.

Discussion

The simulation and modeling of 1-octanol synthesis left a conversion of CO, H_2 and methanol according the reported for Surisetty *et al.*, 2011, Tien-Thao *et al.,* 2007, Hilmen *et al.*, 1998 and other authors (Epling, Hoflund, Hart, & Minahan, 1997; Liu, Shi, Wu, Zhao, & Ren, 2008; Nieskens, Ferrari, Liu, & Kolonko Jr, 2011). It is observed that the 1-octanol yield is influenced for the CO_2 concentration and the pressure (Figures 1 and 2). Higher pressures and low CO_2/CO feed ratio increase the 1-octanol yield. The influence of hydrogen and methanol concentration sees in the Figures 3 and 4 shows that high CH_3OH/CO feed ratio and low H_2/CO feed ratio increase the yield to 1-octanol. If it takes the best operating conditions and design the PBR, results a catalyst charge of 3.9 kg for a yield of 0.043 mole of 1-octanol per mole of CO feeding (Figure 5). This operation condition was taken in the design of the 1-octanol production global process.

The global process was designed from rice husk due to overproduction of rice in Colombia (2’543,161 t per year) (Agronet, 2012) which leaves as residue the rice husk. This residue is transformed in 1-octanol, rice husk ash and electricity with high add value. The yield to 1-octanol is very promising because having potential as jet fuel (see Table 4). However, this potential as jet fuel needs to be evaluated due to high viscosity at -20°C and low freezing point that prevents its direct use in jet motors.

The rice husk ash obtained as subproduct has a good application in the construction industry as biocomposite for its high content of silica that increases the brickwork resistance in the constructions.

Table 4. *Jet fuel physical properties: comparison between Jet A and 1-octanol*

Physical Properties	Jet-A 4658 (Hui, Kumar, Sung, Edwards, & Gardner, 2012)	1-Octanol
Net heat of combustion (MJ/kg)	42.8	38.15
Density at 15°C (kg/L)	0.806	0.834
Viscosity at -20°C (mm^2/s)	4.1	58.16
Flash Point (°C)	47	81
Freezing Point (°C)	-49	-15.5
Mean Boiling Point (°C)	211	193.95
API gravity at 60°F	-	39.1
H/C ratio by mole	1.957	2.25
Molecular weight (g/mol)	142	130.23

The power generation in the process (see Table 2) supplies the 58.75% of energy demand. This result is obtained because the syngas and the combustion gas are used to produce HPV and electricity for the global process. Electricity used reduces the energy requirements and it shows that cogenerated in the process is a good technological option.

The total production cost of a 1kg of 1-octanol obtained was USD 0.957 (Table 3) which in comparison with the sale price (USD 1.46 per kg (Scavage, 2010)) shows profit margin of 34.45% calculated as follows:

$$\text{Profit Margin} = \left(\frac{\text{Sale Price} - \text{Total production cost}}{\text{Sale Price}}\right) * 100 \quad (30)$$

The total production cost distribution (Figure 7) shows that the raw material has the highest impact (78%) due to the methanol need in the 1-octanol synthesis followed the depreciation expenses with a 12% of the total. The mass and energy integration show a considerable influence in the total production cost because to use of recirculation and the fluid distribution around process structure make up for the needs of heating and cooling.

The environmental evaluation shows the total PEI of product leave the system is 0.08142 PEI/kg with a PEI mitigated of 12.97 PEI/kg. This result is very important due to the mass and energy integration in the process mitigates the negative environmental charge of the raw material due to the methanol toxicity and the transform in added value products. The wasted streams have a low environmental impact with an Aquatic Toxicity Potential ATP impact mitigated for aquatic animals if this stream is discharged to the rivers. In the streams leaving at system are observed that HTPI, TTP, PCOP and AP have a low contribution to total environmental impact because for the presence of 1-octanol and H2S traces in the wasted water. The 1-octanol process may be considered environmentally friendly and clean.

Conclusion

The 1-octanol production as a molecule suitable to jet fuel is in discussion because for some properties as the viscosity and the freezing point are not in minimum requirements to jet motor. However, the global production process from rice husk shows a high potential technological, economical and environmental as a sustainable industry at take advantage of an agroindustrial residue and converted in added values products and energy.

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