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PROCESS SIMULATION OF THE PALM OIL HYDROTREATING FOR GREEN DIESEL PRODUCTION USING ASPEN PLUS

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Abstract. Hydrotreated vegetable oil (HVO) also known as green diesel or hydroprocessed renewable diesel, is a biofuel produced from fat matter and obtained from the hydrogenation process. HVO presents similar physical–chemical properties to conventional diesel. Depending on the final properties desired, the fuel can be subjected to an isomerization and cracking process, through the addition of the hydrocracking stage, it is expected the increase in light products. The present work proposes a model for the hydrotreatment of palm oil components for the production of green diesel. The hydrotreating process was analyzed by adding a hydrocracking step and evaluating the changes in the final product in contrast with the process without the cracking stage. Simulations were developed using the Aspen Plus® v.11. It was modeled the main reactions of palm oil in the presence of a NiMo/gAl₂O₃ catalyst in an equilibrium reactor, the model used was Peng Robinson with RK-Aspen. It was analyzed the influence of H₂/oil molar ratio in the process, evaluating the yield of liquid and gaseous compounds. The results showed that decarboxylation reactions (deCO₂) in the hydrotreatment process had a participation of 67.7% in the 12 H₂/oil ratio and decline with the H₂ increase, giving rise to hydrodeoxygenation (OD) that went from 32, 3% to 88.7% for H₂/oil from 8 to 24 respectively. The hydrotreatment of palm oil showed suitability in the production of biofuel using NiMo/gAl₂O₃ catalyst, for low and high H₂/oil molar ratios favoring odd and even hydrocarbons respectively.

Keywords: Renewable diesel, hydroprocessing, palm oil, hydrocracking

1. INTRODUCTION

The rapid grow in energy demand has led to an increase in the fuel consumption, in the same way, along with the need to reductions in the greenhouse gas (GHG) emissions, especially CO₂, are the leading factor to impulse the development biofuels to substitute petroleum-based fuels as diesel and gasoline fuels. Among the substitute fuels for petroleum diesel, there are several renewable options, being biodiesel the most consolidated in production. Biodiesel is less polluting than petrodiesel, has insignificant sulphur content, its biodegradable, and has good lubricating properties, increasing engines' life. Another biofuel substitute for petroleum diesel is green diesel, this is the product of the hydrotreatment process. Due to its superior properties that are quite similar to petroleum diesel, green diesel has become an attractive biofuel.

Another biofuel substitute for petroleum diesel is green diesel, this is the product of the hydrotreatment process. Green diesel, also known as renewable diesel, is a diesel substitute of renewable origin (vegetable oils and fats). It is produced via catalytic hydroprocessing of vegetable oils and fats. Catalytic hydroprocessing is a common refinery process aiming to increase hydrogen to carbon ratio, that use hydrogen to decrease the concentration of heteroatoms such as sulfur, nitrogen, oxygen, and metals, saturate the olefins and aromatics, and to reduce the boiling point of petroleum fractions (Sari, 2013). Catalytic hydroprocessing technology can convert raw vegetable oils, waste cooking oils, animal fats and algal oils into biofuel with high yields. The lipids and triglycerides can be converted into paraffins in the range of gasoline and diesel (Sonthalia; Kumar, 2019). The process is focused on producing a high-quality product that is compatible with existing diesel fuel infrastructure therefore, no further modifications are required to existing infrastructures (Naji, Tye and Abd, 2021).

The Green diesel technology consists of two steps, one catalytic hydrotreatment step which will produce normal paraffins, and one catalytic isomerization or cracking step which will lead to a mixture of n- and iso- paraffins.

(Bezergianni; Dimitriadis, 2013). Afterwards, the products can detach into different components using distillation column on the basis of their boiling point such as gasoline (60–120 °C), kerosene (120–180 °C), and heavy diesel (180–200 °C) (Naji, Tye and Abd, 2021). Fig. 1 shows the different industrial biofuel production processes with the types of raw materials and resulting products. The cetane number of green diesel ranges between 80 and 99 against 45 to 65 observed for biodiesel (Giakoumis; Sarakatsanis, 2019), which is much higher compared to diesel standards, rendering it a competitive diesel substitute. The density range of green diesel is 0.77–0.83 g/ml which also meets the diesel–biodiesel standard. Its net heating value is between 42 and 44 MJ/kg, which is similar to petrodiesel, while its cloud point is from °25 to 30°C. (Bezergianni; Dimitriadis, 2013).

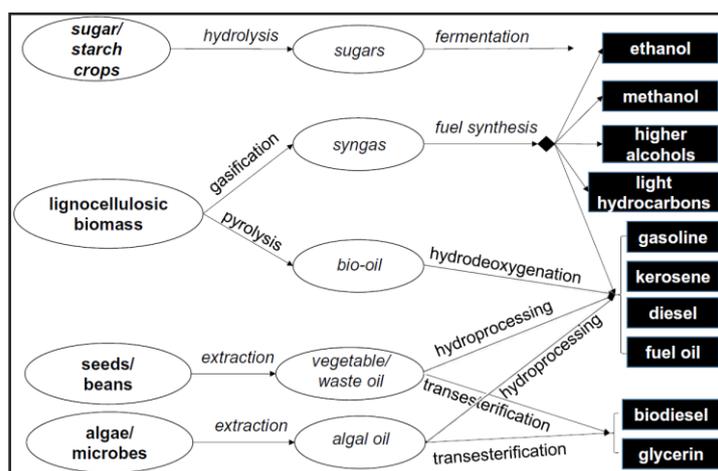


Figure 1. Process pathways for biomass valorization . Fonte: Atsonios et al., 2018.

There are several terms for calling the biobased alkanes/olefins mixture with chemical structure C_nH_{2n+2} , these include “Hydrotreated vegetable oil (HVO)”, “renewable diesel fuel”, “hydrogenation derived renewable diesel”. “Green Diesel” is the trade name given by UOP/Eni whereas company SK Energy refers HVO as HBD (Hydrogen Treating Biodiesel). Hydrotreated vegetable oil (HVO) was first produced commercially by Neste Oil Porvoo Refinery in Finland in the year 2007 plant, with a capacity of 170,000 t per year; with the trade name of NExBTL. Other manufacturers are ConocoPhillips (United States, Ireland), Universal Oil Products (UOP)-Eni (UK, Italy), Nippon Oil (Japan), SK Energy (Korea) and Syntroleum (United States) (Bezergianni; Dimitriadis, 2013; Sonthalia; Kumar, 2019).

The use of several simulation software has been widely extended to study biofuels production, allowing the variation of parameters, reaction models, properties of compounds among others. Recently, Glisic, Pajnik and Orlović, (2016) carried out a technical-economic comparison of HVO and biodiesel using UniSim® software. They compare production methods of biodiesel (supercritical and conventional) with hydrotreatment process, finding Green diesel production by catalytic hydroprocessing located in a petroleum refinery the most cost-effective option.

With the use of SuperPro Designer® Martinez-Hernandez et al., (2019) performed a techno-economic analysis for the production of bio-jet fuel and green diesel in order to find the minimum selling price, in their study they find that because of the bigger minimum selling price of bio-jet fuel, this one would need incentives or subsidies while the green diesel can be competitive with the 2019 fossil diesel prices.

Silva; De Andrade, (2021) used ASPEN PLUS® 8.8 to conduct the conversion of triglycerides by including the rigorous kinetic data of complex reactions, using 32 different types of triglycerides in feedstock, containing a total of 170 species in their model. They found that initial composition of vegetable oil used as feedstock strongly affects the type and hydrocarbons (HC’s) yield. Cavalcanti et al., (2022) simulated the production of green diesel from Brazilian soybean oil considering the hydrocracking step, they assessing the potential of Brazilian soybean oil for the production green diesel, resulting in compliance with quality standards in accordance with chemical properties with the current brazilian legislation for green diesel.

Plazas-González, Guerrero-Fajardo and Sodr , (2018) presented a model of hydrotreating palm oil to produce green diesel using Aspen Plus® 7.3 software to determine the optimized conditions in terms of pressure, temperature and H_2 /oil molar ratio. By means of a packing bed reactor modeled as equilibrium reactor, they predict the behavior reactions of triglycerides and fatty acids in the presence of a $NiMo/gAl_2O_3$ catalyst, they obtained a 97% purity degree of the green diesel and their final product was composed mainly by n-C18 and n-C16 in proportions of 52.9%, 44.2% respectively.

A remaining difficulty now a days for the HVO process is the production costs, due to the high consumption of hydrogen, once this inconvenience is overcome, it is expected that, HVO may surpass biodiesel in technical and environmental aspects, and its production process can also yield aviation fuel. Moreover, since green diesel can be produced from a variety of vegetable oils, a competition for primary resources may also occur (De Souza et al., 2022).

Enhanced isomerization and cracking reaction may produce lighter and isomerized hydrocarbons providing the final product with better flow properties at low temperatures. The present study aims to model the hydrotreating process of palm oil for green diesel production in the presence of a NiMo/ γ -Al₂O₃ catalyst with and without cracking step using the thermodynamic model Peng Robinson with RK-Aspen. Basing the cracking step on a commercial process available under the patent No.: US 9.266,802 B2 (Chen et al., 2016) as used in Lorenzi et al., (2020) work. The simulations were developed using the Aspen Plus® v.11 software.

2. MECHANISM OF HYDROCRACKING OF TRIGLYCERIDES

A pioneer work on hydrocracking of vegetable oils was carried out as early as the 1980s, by Gusmão et al., (1989), they investigated the hydrocracking of soybean and babassu oils for the production of hydrocarbons over sulfided NiMo/ γ -Al₂O₃ and a reduced Ni/SiO₂ catalyst in a batch reactor at temperatures of 350–400 °C and hydrogen pressures of 1–20 MPa obtaining a mixture of hydrocarbons, essentially normal alkanes. Despite all the advances in the hydrotreating process mechanism and kinetics, these are still under investigation because of its complexity. Since most vegetable oils usually contain high values of triglycerides (80-90%), the development of reaction mechanism focuses on the hydrocracking of triglyceride molecules. During the hydrocracking process, the richness of hydrogen in the reactor saturates the side chains of the triglycerides, so double bonds will be converted to single bonds. This is then followed by scission of C-O bonds to produce free fatty acids. Then free fatty acids are subjected to three main reactions: hydrodeoxygenation (HDO), the hydrodecarbonylation (deCO), and the hydrodecarboxylation (deCO₂). In order to remove oxygen. Oxygen must be removed to obtain a liquid fuel with a high-energy content and high thermal stability similar to fossil fuels. The produced straight chain alkanes may undergo isomerization and cracking reaction. The isomerization reaction is responsible for converting straight chain alkanes to branched chains to enhance the energy content and prevent engine knocking. The cracking reactions will convert long chain hydrocarbons to shorter chains in order to improve the selectivity to diesel, jet fuel, and gasoline (Alwan, 2014).

2.1 Hydrotreatment Reaction Route

The main hydrotreatment reaction route is illustrated in Figure 2. The first reaction in the hydrotreating process is saturation of double bonds present in triglycerides. Each double bond requires one H₂ molecule. The introduction of excess hydrogen in the hydrotreatment process helps to break the C=C double bond present in the triglycerides, obtaining single bonds. At the next reaction step the triglycerides are cleaved at the C-O bond leaving one propane molecule and three fatty acids. Cracking ensures that the chain length of the product is within the boiling point range of gasoline, diesel, and kerosene. The presence of oxygen in the final product reduces oxygen stability and heating value of bio-based feedstock, and it also increases the acidity and corrosivity. Thus, its removal is necessary (Sonthalia; Kumar, 2019). The oxygen removal and conversion of the fatty acids to hydrocarbons follows three major pathways: The HDO, deCO, the deCO₂. The HDO pathway consumes the most hydrogen but does not reduce the length of the carbon chain, and, thus, maximizes the green diesel yield. Noteworthy, the only byproduct is water. The deCO reaction consumes only a third of the hydrogen but has the disadvantage of removing one carbon atom from the carbon chain in the form of CO. Lastly, the deCO₂ pathway requires no hydrogen at all and removes the two oxygen atoms as CO₂. Depending on the catalyst and the reaction parameters various side products like diglycerides, monoglycerides, olefins, esters, and alcohols can appear. Also cracking and isomerization of the hydrocarbons are a possibility, resulting in short-chained or branched hydrocarbons, respectively. In the gas phase CO and CO₂ can be converted into each other following the water gas shift reaction (Kiefel; Lüthje, 2018).

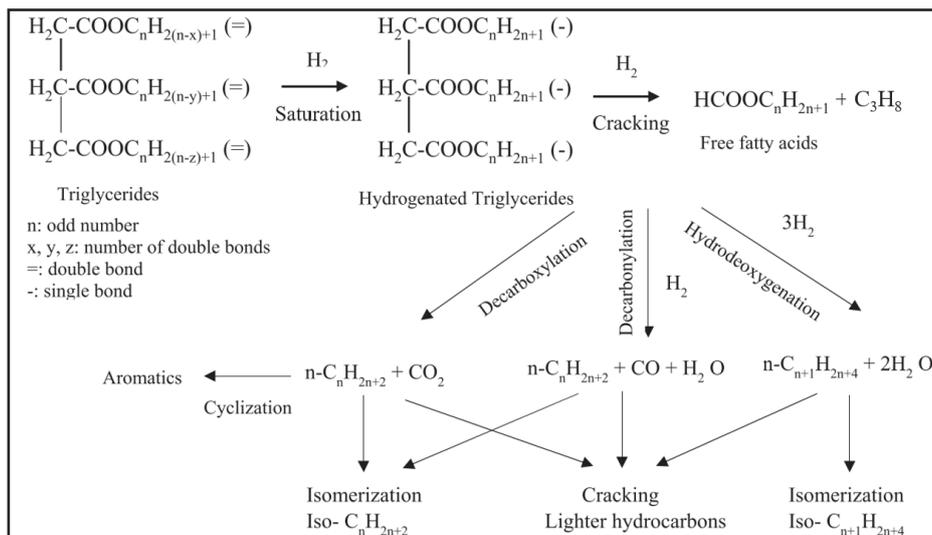


Figure 2 The three main hydrotreatment pathways. (Sonthalia; Kumar, 2019)

The isomerization reaction is responsible for converting straight chain alkanes to branched chains to enhance the energy content and prevent engine knocking. The cracking reactions will convert long chain hydrocarbons to shorter chains in order to improve the selectivity to diesel, jet fuel, and gasoline (Alwan, 2014). Different parameters, such as raw material composition, activity and operating conditions, directly affect the final properties of hydrocarbon biofuels. Products with low oxygen content and shorter chains exhibit weaker molecule interactions, which correspond to low viscosity and density properties. In the same context, catalytic cracking promotes the removal of oxygen atoms increasing the higher heating value to approximately 45 MJ/kg. Meanwhile the cetane number of the direct organic liquid product from the catalytic cracking of vegetable oils can be in between 70–90, since product with high cetane number reduces the ignition time, resulting in high-efficiency ignition (Naji; Tye; Abd, 2021). In general, the catalysts that are used in the hydrocracking of vegetable oils should consist of two sites: metallic sites and acidic sites. The metallic sites are required for hydrogenation, HDO, deCO, the deCO₂ reactions. The acidic sites are required for isomerization and cracking reactions. Therefore, catalyst design needs an equilibrium between metal and acid sites, in order to modify selectivity, activity, and durability of the catalysts (Alwan, 2014). According to Veriansyah et al., (2012) enhanced isomerization and cracking reaction activity on the CoMo catalyst may produce lighter and isomerized hydrocarbons.

3. HYDROTREATMENT AND CRACKING SIMULATION

Aspen Plus® 11 software was used to model biofuel production by means of palm oil hydrotreating. To simulate the main triglycerides reactions: fatty acid hydrogenation, decarboxylation, reduction, alcohol formation and cracking, the thermodynamic method used to model the conditioning of reactants was Peng-Robinson with RK-Aspen, as it fits to hydrocarbons processing and its able to predict at high pressure the behavior of gas-liquid equilibrium of triglycerides. The triglycerides composition of palm oil to feed stream to the reactor is 37% triolein, 48.6% tripalmitin, 10% trilinolein and 4.4% tristearin (Plazas-González; Guerrero-Fajardo; Sodr , 2018). The catalyst used in the hydrotreating model was 5%Ni15%Mo/γ-Al₂O₃, with an oil (kg/h)/catalyst (kg) ratio of 30:1. As NiMo, is considered a promising concerning selective deoxygenation for producing green diesel of natural triglycerides according to Kordulis et al., (2016).

A simplified diagram of the simulation is Fig. 1. The simulation consists of a feed oil and hydrogen which is preheated and fed to the hydrodeoxygenation reactor (B3) which operates at 360 °C, 40 bar, according to experimental work of (Kiatkittipong et al., 2013) on oil palm hydroprocessing, in which diesel yield increases to the maximum value at 40 bar and then levels off at higher pressure. The reactor outlet is a mixture of linear saturated hydrocarbons, and gases including mainly water, CO, CO₂, alcohols and propane.

The outlet stream of the hydrotreatment reactor firstly goes through a flash tank (FLASH1), operating at 70 °C, which is used to separate liquid hydrocarbons from water and syngas. The liquid hydrocarbons goes to a second low pressure, low temperature tank (FLASH2) that depressurizes the blend and volatilizes the gas for separation, obtaining hydrocarbons in the bottom, n-C15 to n-C18. The vapor stream from the first flash tank (FLASH1) and the second tank (FLASH2) flows through a membrane (MEMBRANE) with a hydrogen selectivity of roughly 100% and the resulting stream is recirculated to join initial hydrogen fed. Low-cost, high-temperature hydrogen separation membranes represent a key enabling technology for small-scale distributed hydrogen production units. The availability of such membranes with high selectivity and high permeability for hydrogen enables their wide applicability with hydrocarbon reforming and water gas shift (WGS) reactions, potentially reducing the cost of hydrogen produced (Pall Corporation; Colorado School Of Mines, 2012).

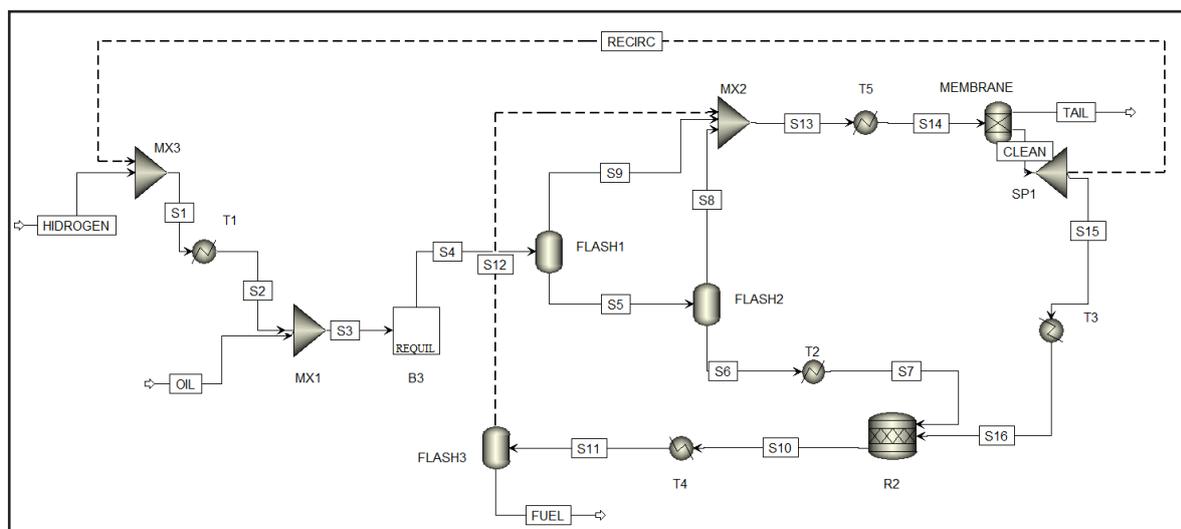


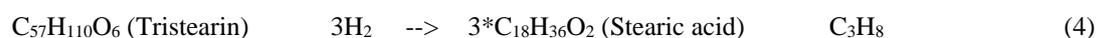
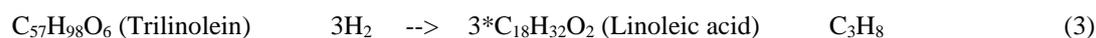
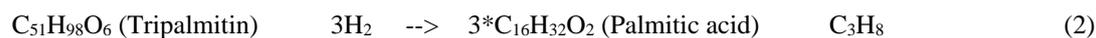
Figure 3. Simplified process flowsheet of palm oil hydrotreatment in Aspen Plus.

In the case of the green diesel hydrodeoxygenation including isomerized stage, the process is modified by a second catalytic isomerization step to convert the linear paraffins into iso-paraffins, this stage is mainly done to improve the biofuel cold flow properties (Del Río et al., 2021). The liquid stream reaches 355 °C and 50 bar and it is directed into R2 with the function of partially isomerizing the n-alkanes. R2 was simulated as a stoichiometric reactor considering simplified mass yield basis, 10% of both C₁₇H₃₆ and C₁₈H₃₈ was assumed to be isomerized into 6-pentyldecane and 8-propylpentadecane respectively, and 4.7% wt heptadecane is cracked into propane (Chen et al., 2016; Letterio; Mcnew, 2016; Lorenzi et al., 2020)

In the hydrotreatment process simulation, H₂ to oil molar ratio was varied from 8 to 24 in order to evaluate its effect on the final product in both processes and the weight of HDO, deCO, the deCO₂ reactions in the palm oil hydrogenation.

Table 1. Reaction pathways involved in conversion of triglycerides into hydrocarbons. (Veriansyah et al., 2012)

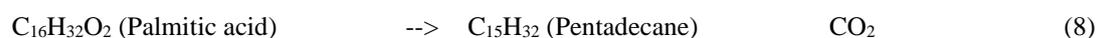
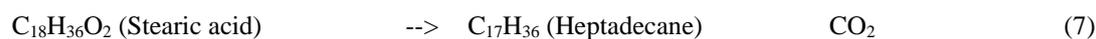
INITIAL REACTIONS



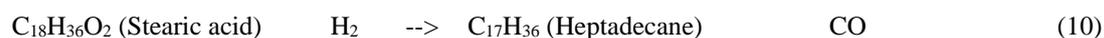
HYDROGENATION

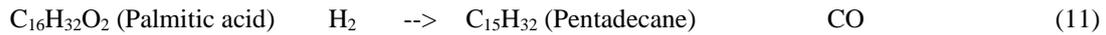


DECARBOXILATION (deCO₂)

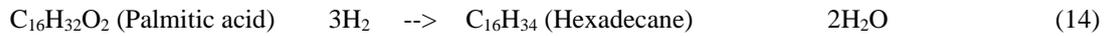
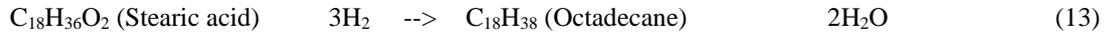


DECARBONILATION (deCO)

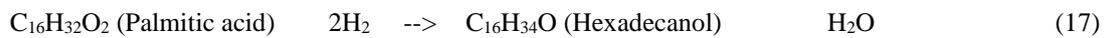




HYDRODEOXYGENATION (HDO)



ALCOHOL FORMATION



From Tab. 1 it can be observed that HDO reaction produces hydrocarbons with even number of carbons Eq. (13 to15). While the *deCO* Eq. (10 to12) and *deCO₂* Eq. (7 to 9) reactions produce hydrocarbons with odd number of carbons. These allowed calculating the contribution of each reaction to the product distribution involved hydrotreating process. For example, if the mass of produced hydrocarbons, CO and CO₂ are measured, the percentage of each reaction type can be calculated from the following set of equations (Martinez-Hernandez et al., 2019).

$$\frac{\sum m_{even}}{\sum m_{odd}} = \frac{HDO}{deCO+deCO_2} = A \quad (18)$$

$$\frac{m_{CO_2}}{m_{CO}} = \frac{deCO_2}{deCO} = B \quad (19)$$

$$HDO + deCO_2 + deDCO = 1 \quad (20)$$

Where *m_{even}* is the mass of hydrocarbons with even carbon number, *m_{odd}* is the mass of hydrocarbons with odd carbon number, while *m_{CO₂}* and *m_{CO}* is the mass of CO₂ and CO produced, respectively. This HDO, *deCO*, the *deCO₂* reactions are the percentage or extent of the reaction taking place and contributing to the product distribution.

4. RESULTS

After configuring all process inputs, the steady-state simulation was completed without convergence errors. The H₂/oil molar ratio was varied from 8:1 to 24:1.

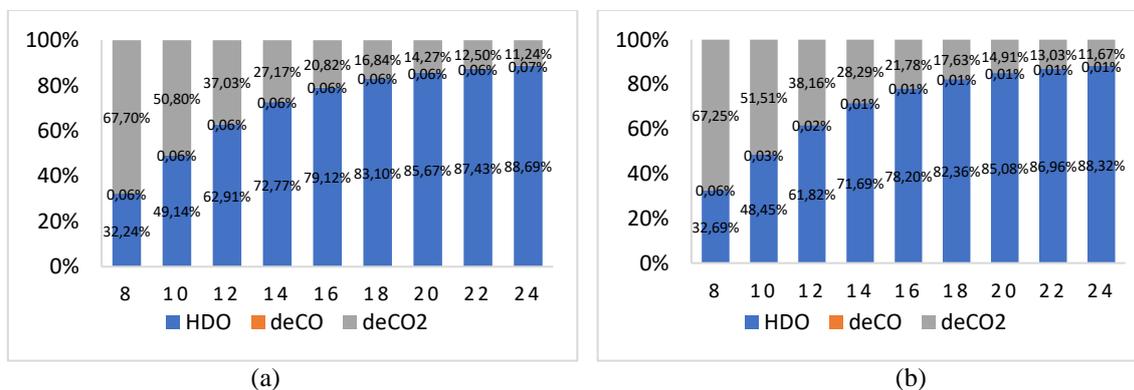


Figure 4. Results of extent of The HDO, *deCO*, the *deCO₂* reactions for the simulation of green diesel production for simple process (a) and process including cracking step (b).

As the H₂/oil molar ratio increases, the HDO reaction contribution, increases and deCO₂ reaction decreases. This behavior was appreciated for the process with and without hydrocracking (Fig. 4). The contribution of HDO increases along with the H₂/oil ratio until it contributes to 88% of the biofuel formation in the relation of 24, H₂ to palm oil molar rate.

The presence of H₂ in the reaction atmosphere leads to improvements in the deCO reaction in the simple process, tending to favor the HDO pathway, although the values are low (<0,07%). In the case of cracked green diesel production, the contribution of deCO decreases to almost zero in the reaction, reaching the value of 0.01% in the relation of 24 H₂ to oil molar rate. The deCO₂ reaction had a higher percentage share in the hydrocracking process at low H₂ values, for both process.

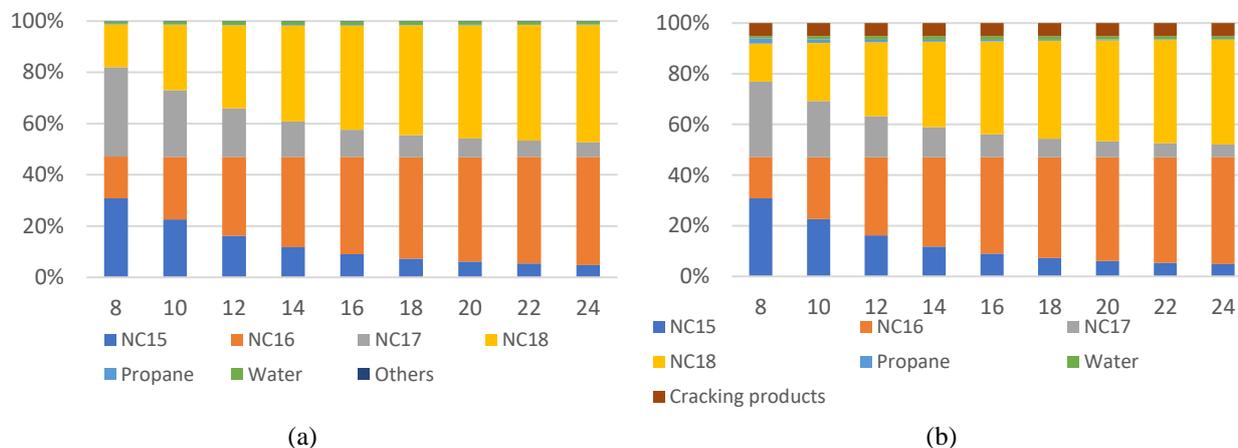


Figure 5. Effects of H₂/oil molar rate on the mass fraction and mass flow rate of the liquid products for simple process (a) and process including cracking step (b).

With increasing H₂/oil ratio, the production of n-C16 and n-C18 compounds, are increased. In the hydrotreatment of palm oil, high amounts of hydrocarbons were obtained in the final product, reaching up to 98% of the final product (Fig 5). For lower H₂/oil ratios, the production of HC's n-C15 and n-C17 was dominant, contrary to the production of n-C16 and n-C18, which had a higher formation from H₂/oil 10 ratio onwards. Recalling that working with low H₂/oil ratios, its possible to obtain high fractions of HC's but at expenses of the rapid deactivation of the catalyst. It is well known that rapid deactivation of catalysts occurs in the absence of hydrogen, requiring constant catalyst regeneration. (Kordulis et al., 2016) and reduction in pressure or H₂/feed oil ratio could lead to increased oligomerization reactions providing a route to more rapid catalyst deactivation as experimeted in the Anand and Sinha (2012) work, for this is necessary to consider working with high H₂ to minimize oligomerization of the products and to increase the lifespan of the catalyst.

The cracking step allowed obtaining up to 5.17% in the final product isomers, this fraction consisting basicalli of 6-pentyldodecane and 8-propylpentadecane. In the final flow of HVO, a water share of up to 1.5% was obtained, an additional dehydration step may be necessary for its removal.

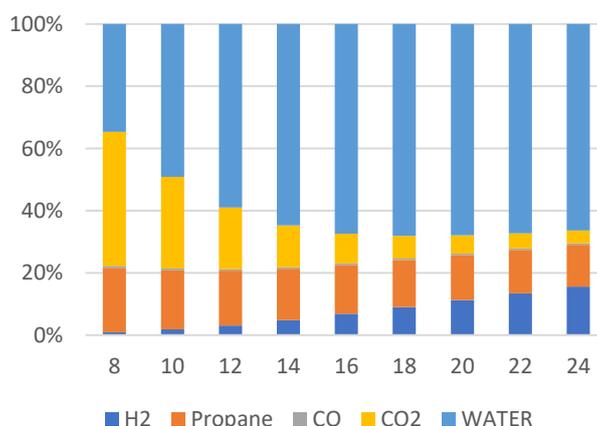


Figure 6. Effects of H₂/oil molar rate on the molar fraction of gaseous products.

The Fig.6 shows the output gases once the H₂ recovery takes place. Up to 67% of the output gases of the process are composed of water vapor, from 34% reaching up to 67%. This is in agreement with the higher participation of HDO reaction shown in figure 1. For reduction equations (13-15), every three moles of H₂ reacting with a free fatty acid produces two moles of water and one hydrocarbon.

For higher H₂/oil values, production of propane decreases from 20% to 13%. The production of propane and CO₂ is notable for low H₂ ratios reaching 62% of the total mass composition of the gases. Once the H₂/oil ratio reaches the value of 18, the sum of propane and CO₂ acquires an approximate value 20%. The composition of the output gases of the two processes showed an identical composition. The variation of H₂/oil present a minimum impact in the production of CO, its values stay from 0,65 to 0,51%, in accordance whit the participation of deCO reaction.

5. CONCLUSION

The simulation and analysis of the production of green diesel from hydrotreatment of palm oil was presented in this paper. The biofuel process involved hydrodeoxygenation and hydrocracking to yield green diesel. A packing bed reactor modeled as equilibrium reactor was used to analyze the H₂/oil molar ratio the deoxygenation of vegetable oils for production of green diesel using Aspen plus.

As the H₂ to oil molar ratio increases, the most favored reaction is hydrodeoxygenation (HDO). With the catalyst 5%Ni15%Mo/ γ -Al₂O₃ it was possible to obtain high values of HC's n-C15 to n-C18, being for smaller H₂/oil the production of odd HC's and for H₂/oil bigger to 12 greater the production of even HC's . The simulation of the isomerization reactor through the stoichiometric model allowed to obtain hydrocarbons with shorter chains following the information contained in the patents, obtaining a share of between 5.13 and 5.17% of 6-pentyldodecane and 8-propylpentadecane in the final product. Hydrocracking is a way to obtain lighter HC's and improve cold flow properties.

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