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SIMULATION AND OPTIMIZATION OF CONTINUOUS BIODIESEL PRODUCTION FROM SOYBEAN OIL

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Abstract. *It is known that oil and its derivatives were the main energetics of the last century, although the fact that these inputs are finite and generate several pollutant emissions. Economic and environmental aspects have contributed to the gradual disuse of fossil fuels and to the growth of research on renewable and clean energy, such as biodiesel, that has been gaining representativeness in the national and world market. Brazilian biodiesel production has been led by soybean oil for many years, then, aiming to make the biodiesel plant (based on biodiesel process described by Zhang et al., 2003) the raw material (triolein) was changed to (trilinolein) through the use of the predictive method of Constantinou e Gani (1994) to estimate base and critical properties. Besides, a kinetic equation based on the experimental data of Noureddini and Zhu (1997) was proposed to represent the rate of soybean transesterification in function of temperature. Finally, optimization studies were made with the objective of maximizing biodiesel production; minimize the consumption of energy and inputs; outline great scenarios from the environmental perspective*

Keywords: *Biodiesel, Simulation, Optimization, UniSim, Environment*

1. INTRODUCTION

The advance of the automobile industry in the twentieth century raised oil and its derivatives to the level of the principal representative of the world energy matrix (FARIAS & SELITTO, 2011). However, economic and environmental aspects have contributed to the disuse of these fuels, such as the oil crises of 1973 and 1979 that caused an increase in the prices of barrels with consequent destabilization of the world economy (MME, 2017).

Also, fossil fuels are non-renewable sources of energy that produce emissions of pollutants such as carbon dioxide and particulate matter, harmful to human health and the environment (SILVA, 2012).

At the turn of the 21st century, a greater environmental awareness began, motivated by studies on the impacts of the anthropological action on the environment and on the effects that such actions would bring to the ecosystem as well as the quality of life of the next generations (TESSMER, 2002). Thus, there is a global incentive to research and the use of clean and renewable energy in order to modify how natural resources are exploited.

There is now a gradual change in the world energy matrix. Concerning biomass, it is important to stress that the one of liquid origin, commonly known as biodiesel, has been gaining representativeness in both national and world market. The main producers and consumers of biodiesel are the USA, Brazil and some European countries, according to the Renewable Energy Policy Network for the 21st Century (REN 21).

Biodiesel can be produced from three groups of raw materials: oils (edible and inedible), fats (bovine, pork, chicken) and waste material (frying oil, other fatty materials, etc.). Soybean oil is the most widely used crop in the world, especially in leading countries in biodiesel production. This is due to its large-scale cultivation and the ability to adapt to different climates (ALVARÃES, 2017).

Among the production routes of this biofuel, the one that involves the transesterification is the most used. In this process, triglycerides present in the oil react with a short-chain alcohol (usually methanol or ethanol) in the presence of a catalyst, producing a mixture of esters, commonly known as biodiesel, and glycerol (GERPEN *et al*., 2004; FERRARI, 2005; SANTANA, 2008). Several studies have been carried out to determine the kinetics of transesterification reactions between oils and alcohols. In general, for molar ratios of about 6: 1 for alcohol / oil, a second-order kinetics is assumed (NOUREDDINI & ZHU, 1997).

This work made a survey of the latest research on biodiesel production (Table 1) that showed that few studies are devoted to optimization of biodiesel plants using simulation tools.

Table 1. Recent research on biodiesel production

Authors	Simulator	Process	Energy Analyis	Economic Analysis	Optima zation
West *et al*. (2008)	Hysys®	Alkali or continuous acid-catalyzed	Yes	Yes	No
Pokoo Aikins *et al.* (2010)	ICARUS®	Alkali-catalyzed	Yes	Yes	No
Santana *et. al* (2010)	Hysys®	Alkali-catalyzed	Yes	Yes	No
Lee *et al.* (2011)	Hysys®	Alkali-catalyzed	Yes	Yes	No
Nguyens & Demirel (2013)	Hysys®	Acid-catalyzed	Yes	Yes	No
Coronado *et. al* (2013)	Hysys®	Alkali-catalyzed	Yes	Yes	No
Coronado *et. al* (2014)	Hysys®	Alkali-catalyzed	Yes	Yes	No
Plate *et al.* (2014)	IFSH	Alkali or Continuous Acid-catalyzed	Yes	No	No
Maceiras *et. al* (2016)	iQuimicaCAD®	Continuouswith diferentes feedstocks	No	Yes	No
Alvarães (2017)	SimSci PRO/II®	Alkali and Enzyme-catalyzed	Yes	No	Yes

Thus, the main objective of this paper is to propose the optimization of a continuous biodiesel plantaiming to maximize biodiesel production and purity. In order to carry out this study, the methodology of the work will be divided into the following stages:

(a) Reproduce and validate the process proposed by Zhang *et al.* (2003) using Honeywell UniSim R390 software.

(b) Propose a modification in the transesterification reactor, from fixed conversion to a kinetic equation based on experimental studies.

(c) Process optimization using the UniSim optimization toolbox.

2. METHODOLOGY

2.1 Validation of the present work with data provided by Zhang *et al.* (2003)

The authors developed the continuous biodiesel production process through the HYSYSsimulator. However, this work usedthe Honeywell Ltd.'s UniSim Design R390 simulator due to the ease of access to this software and its similarity to HYSYS.In Fig.1 it is represented the PFD (process flow diagram) of the continuous biodiesel process.

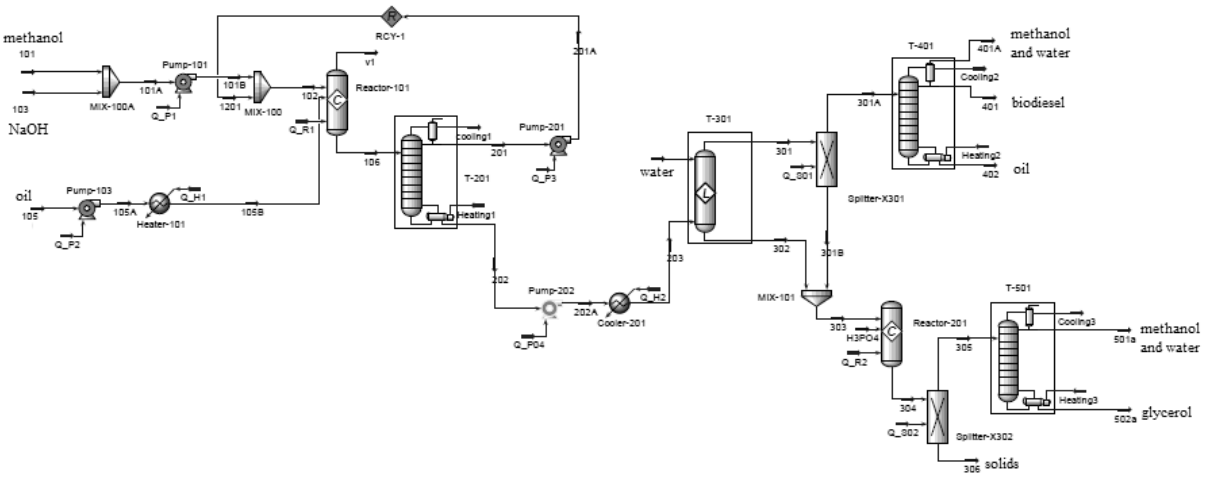


Figure 1 - Continuous biodiesel production process

The biodiesel production process studied in this paper is based on the plant proposed and simulated by Zhang *et al.* (2003). The continuous production of biodiesel is carried out by the methyl transesterification of soybean oil in which sodium hydroxide is used as a catalyst. The compounds used in this simulation were: triolein, diolein, monolein, sodium hydroxide, water, phosphoric acid, sodium phosphate, methanol, glycerol and methyl oleate.

Basically, the methanol at a flow rate of 3,658 kgmol / hr is mixed at a flow rate of 1,194 kgmol / hr of oil and is fed into the transesterification reactor, R101, where 95% of the oil is converted to biodiesel. The mixture is then sent to a distillation tower, T-201, where methanol is recycled at the top, at a molar flow rate of 3,652 kgmol / hr and the remainder exits from the bottom at a flow rate of 5,090 kgmol / hr. The bottom mixture goes to an extraction tower, T-301, where water is injected at the top, at a flow rate of 0.6106 kgmol / hr, extracting most of the glycerol from the bottom. At the top of the extraction tower the biodiesel-rich mixture, with 3,553 kgmol / hr, goes to the distillation tower, T-401, where the biodiesel is purified at the top, at a flow rate of 3.4 kgmol / hr and a molar fraction of 0.996. The bottom of the extraction tower is sent to an NaOH neutralization reactor (catalyst) and followed to a

distillation tower, T-501, where the glycerol is separated at the bottom, at a flow rate of 2077 kgmol / hr. A detailed description of the plant may be encountered in Zhang *et al.* (2003).

Some pure compounds adopted by Zhang *et al.* (2003) such as triolein, phosphoric acid (H_3PO_4) and sodium phosphate (Na_3PO_4) are not present in the UniSim library. The use of the HypoManager tool allows the addition of these components to the process by inserting the following thermo physical data: molar weight (MW), density (d), normal boiling temperature (T_b), critical temperature (T_c), critical pressure (P_c), critical volume (V_c) and accentric factor (ω).

It is worth mentioning that the HypoManager tool can create a molecule only with the insertion of base data (MW, d, T_b) and estimate the other properties with the use of the Estimate Unknown Props option. Zhang *et al.* (2003) made use of this tool capacity and used basic data contained in Zhang (2002) for triolein - molar weight of 884.00 g/mol; normal boiling point of 500.00°C and a density of 900.00 kg/m^3.

The same procedure was used for phosphoric acid and sodium phosphate, but without specifying the values adopted. Thus, this work inserted base data found in the literature for these compounds - Table 2 - and estimate the remaining data.

Table 2. Base data used for phosphoric acid and sodium phosphate

Molecule	**Formula**	**MW(g/mol)**	**d (kg/m³)**	**T_b (°C)**
Phosphoricacid	H_3PO_4	97.99	1885.00	158,00
SodiumPhosphate	Na_3PO_4	163.94	2536.00	-

Due to the highly polar characteristics of methanol and glycerol and following the procedure described by Zhang *et al.* (2003), the NRTL extended thermodynamic package was chosen to predict the activity coefficients of the components in the liquid phase.

The binary parameters were estimated using the liquid-liquid equilibrium modulus UNIFAC, while the methanol / triolein pair had their activity coefficient estimated by immiscible UniSim tool.

With the use of Excel® from Microsoft, it was possible to construct comparative tables to validate the process simulated by the present work with the data provided by Zhang *et al.* (2003).

2.2 Change from canola oil to soybean oil: from triolein to trilinolein

Canola oil is the main plant material used in the biodiesel generation plants of Canada. The work of Zhang *et al.* (2003) considered this raw material for simulation of a continuous biodiesel plant. However, such raw material does not represent the reality of Brazilian biodiesel production, led by soybean oil for many years. Although triolein plays a significant role in the composition of soybean oil (about 23,26%) trilinolein accounts for more than half of the triglycerides of the mixture (about 55,53%) (GERPEN *et al*., 2004; VENDRAMIN, 2010; ARVELOS, 2013).

Thus, aiming to bring the biodiesel production process of Zhang *et al.* (2003) closest to the national reality, the representative molecule of the oil used as a reagent in the transesterification reaction was altered from triolein ($C_{57}H_{104}O_6$) to trilinolein ($C_{57}H_{98}O_6$). Consequently, the biodiesel generated was composed of methyl linoleate ($C_{19}H_{34}O_2$).

Trilinolein is not present in the UniSim library, so HypoManager was once again used to create this molecule. Although this tool can estimate the critical properties and the accentric factor only with the addition of the base data (MW, d, T_b), the use of accurate information provides results that are closer to reality. The molar weight and densities of trilinolein and were found in the literature with the values of 500.00 g/mol and 900.00 kg/m^3, respectively.

The present work maintained the use of Hypo Manager's Estimate Unknown Props option for phosphoric acid and sodium phosphate. With regard to trilinolein, a comparative study was carried out on the literature of the recommended predictive methods for the calculation of the thermodynamics properties necessary for the development of a hypothetical molecule in the UniSim, emphasizing the absence of T_b experimental data, as shown in Table 3.

Table 3. Comparison of the predictive methods recommended by the literature for calculations of the data necessary to create a hypothetical molecule in UniSim

Data	Poling*et al.* (2001)	Vendramin (2010)	Glisic&Skala (2009)	Arvelos (2013)	Silva (2016)
T_b	-	Experimental	CG (1994)	CGou MG	CG (1994)
T_c	CG (1994)	Ambrose (1980)	CG (1994)	MG (2001)	CG (1994)
P_c	Allmethods	Ambrose (1980)	CG (1994)	MG (2001)	CG (1994)
V_c	Allmethods	Ambrose (1980)	CG (1994)	Allmethods	CG (1994)
ω	CG (1995)	Edmister (1958)	CG (1995)	-	-

In this way, as recommended by a large part of the analyzed literature, the present work adopted the Constantinou & Gani (1994) method to calculate the normal boiling temperature (T_b), critical temperature (T_c), critical pressure (P_c), critical volume (V_c) and the Constantinou & Gani (1995) method to calculate the accentric factor (ω) of trilinolein.

Eq.1 represents the general form of the both CG model (1994) and (1995), where F is the desire function; N_k and M_j are the number of First-Order groups of type "k" and the number of Second-Order groups of type "j", respectively; F_{1k} and F_{2j} are the contribution for the First-Order group named "1k" and F_{2j} is the contribution for the Second-Order group named "2j", respectively.

$$F = (\sum_k N_k \cdot F_{1k}) + W \cdot (\sum_j M_j \cdot F_{2j}) \quad (1)$$

This method is based on the UNIFAC groups and is more accurate due to the consideration of second-order groups contributions that allow the differentiation of molecules with peculiarities such as: resonance structures, isomers, identical chemical groups, etc. There are no such structures in the trilinolein molecule, so W = 0 was adopted. In Table 4 are the values of the CG (1994) and (1995) methods for several chemical groups:

Thus, the Eq. (2)-(6) represents the equations for the calculation of the desired properties:

$$T_b = 204.359 \cdot \ln(\sum N_k \cdot T_{bk}) \quad (2)$$

$$T_c = 181.28 \cdot \ln(\sum N_k \cdot T_{ck}) \quad (3)$$

$$P_c = (\sum N_k \cdot P_{ck} + 0.10022)^{-2} + 1.3705 \quad (3)$$

$$V_c = -0.00435 + (\sum N_k \cdot V_{ck}) \quad (4)$$

$$\omega = 0.4085 \cdot [\ln\left(\sum_k N_k \cdot \omega_{1k}\right)]^{\left(\frac{1}{0.5050}\right)} \quad (5)$$

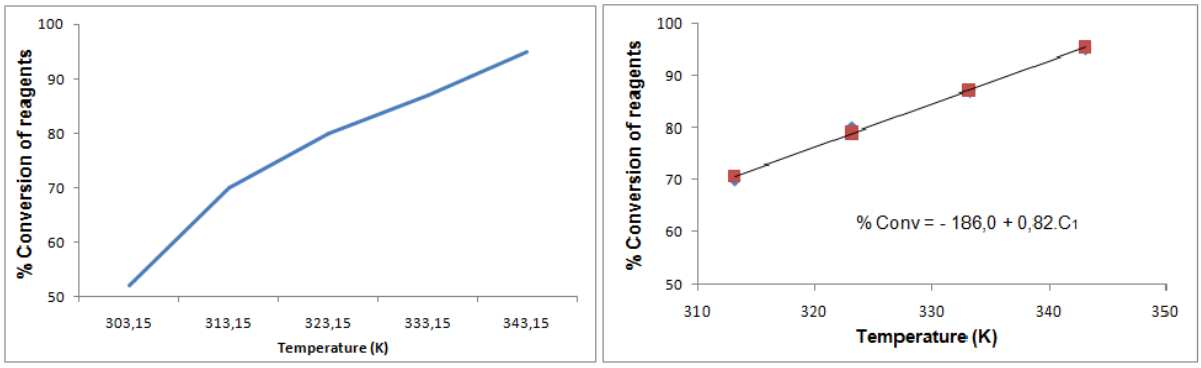
Thus, the calculation of the properties becomes: T_b equals to 827.94 K; T_c equals to 976.8K; P_c equals to 3.37bar; V_c equals to 3.21m^3/ kmol; ωequals to 2.02.

Furthermore, this work maintained the thermodynamic package NRTL Extended to predict the activity coefficients of the components in the liquid phase and the liquid-liquid equilibrium modulus UNIFAC to estimate the binary parameters, except for the methanol / trilinolein pair that had its activity coefficient estimated by immiscible UniSim tool.

In order to validate the results, themainthermodynamic variables of the principal process streams were compared o the results reported by Zhang *et al*. (2003).

2.3 Alteration of the fixed-conversion reactor of the biodiesel production plant described by Zhang *et al.* (2003)

Noureddini and Zhu (1997) studied the kinetic behavior of the transesterification reaction between soybean oil and methanol using sodium hydroxide as an alkaline catalyst. With the Anova data analysis tool, present in Excel, the linear regression of the data related to the conversion of the reactants to different temperatures, set time of reaction and degree of agitation (Reynolds number) of the mixture was performed. Figure2 represents the curve obtained experimentally and Figure 3 the curve after the use of linear regression.



Figures 2 and 3. Experimental conversion data for different temperatures and linear regression curve of the data present in Noureddini& Zhu (1997)

Thus, the 95.00% fixed conversion previously used was withdrawn in order to make the process more realistic. The equation provided by linear regression was added to the reactor where transesterification occurs, obtaining 87.18% conversion of triglycerides

$$C = 0.82T - 186.0 \quad (6)$$

2.4 Optimization of the biodiesel production plant described by Zhang *et al.* (2003)

The introduction of the equation obtained by linear regression resulted in a decrease in the mass flow produced from biodiesel. Thus, aiming at a more efficient process, with lower energy cost and environmentally favorable, a series of optimizations was performed with the help of the UniSim Optimizer tool.

The basic formulation of the problem is based on the maximum biodiesel outflow, as present bellow:

$$max_{\mathbf{x}} f(x) = max_x(\dot{m}_{Biodiesel}) \quad (1)$$

$$h_j(\mathbf{x}, \mathbf{y}) = 0; \quad j = 1, \dots, m_e \quad (2)$$

$$g_j(\mathbf{x}, \mathbf{y}) \leq 0; \quad j = 1, \dots, m_i \quad (3)$$

$$\mathbf{x}_{min} \leq \mathbf{x} \leq \mathbf{x}_{max} \quad (4)$$

Where f, g and h refers to objective function, inequality constraints and equality constraints, respectively, m_i refers to the number of inequality constraints while m_e refers to the number of equality constraints. In addition,x_i, $i = 1 \cdots n_x$ are the decision variables and n_x is the number of decision variables. Finally, y represents all the other state variables.

Table 5 presents the decision variables adopted (X_i) and their respective maximum and minimum constraints.

Table 5. Decision variables adopted and their respective maximum and minimum constraints

Object	X_i	$X_{i\ min}$	Current Value	$X_{i\ max}$
106	Temperature	60.00	60.00	73.00
Water	Mass Flow	5.00	11.00	15.00

3. RESULTS AND DISCUSSION

3.1 Results of the comparison of simulation of these work with the data provided by Zhang *et al.* (2003)

The simulated results were very close to those provided by the authors, allowing the validation of the simulation and follow-up to the change of the oil used as raw material.

3.2 Result of the introduction of the fixed-conversion reactor based on the experiment of Noureddini and Zhu (1997)

The change in the fixed conversion reactor (95.00%) resulted in a transesterification reaction with an efficiency of 87.18%. Comparing the biodiesel production obtained by Zhang *et al.* (2003)withthe obtained by the simulation of this paper, before and after the change of the use of the parameterized equation, shows a decrease in the molar and mass flow rates of biodiesel.

However, an analysis of the composition of the biodiesel in stream 401 evidences the permanence of high purity, around 99.9%. Table 6 shows such comparisons, where 401 represents the values of Zhang *et al.* (2003), 401* represents the simulated results before the

change in the fixed conversion reactor and 401** the results after the use of the parameterized equation.

Table 6. Comparison of the biodiesel production data obtained by Zhang *et al.* (2003) with the data simulated by this dissertation, before and after the change of the fixed conversion reactor

Stream	401	401*	401**
Molar Flow(kgmol/h)	3.38	3.37	3.09
Mass Flow (kg/h)	999.88	999.88	915.81
Mass Fraction of Biodiesel	0.997	1.000	1.000

3.3 Result of the plant optimization aiming the maximization of biodiesel flow rate

The results of the maximization of biodiesel mass flow rate can be visualized in the Table 7 below. The table's columns list the optimizer iterations, number of objective function evaluations, objective function, reactor temperature and mass flow rate of water used the extractive tower.

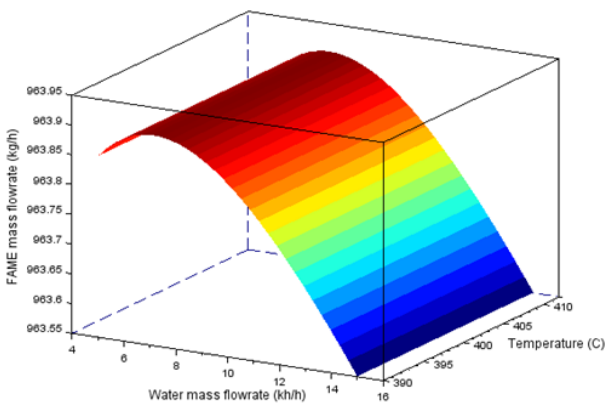
Table 7. Results of the maximization of biodiesel mass flow rate

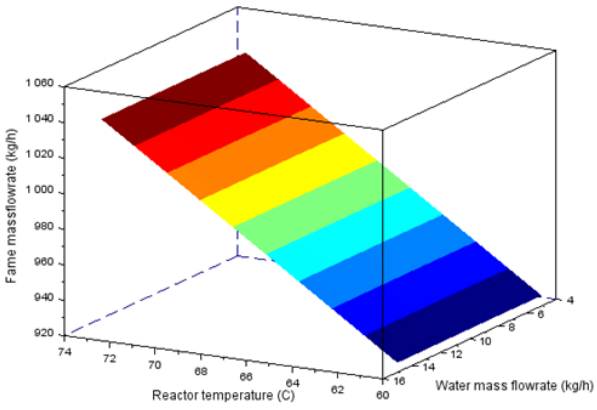
Iteration	Number of Objective Function Evaluations	Objective Function	Temperature (°C)	Water flow rate (kg/h)
5.00	9.00	1038.26	73.00	7.33
4.00	8.00	1038.26	73.00	7.33
3.00	7.00	1037.80	73.00	10.48
2.00	5.00	970.99	65.88	7.85
1.00	3.00	954.36	63.93	11.00

The results indicate that five iterations were sufficient for the algorithm convergence. It can be observed in Table 7 that the maximum biodiesel mass flow rate was 1038.26 kg/h. As indicated, the optimal reactor temperature corresponded to 73 °C and the optimal water flow rate was 7.33 kg/h. Notice that the optimal temperature is exactly the maximum bound configured in the optimization procedure, however the optimal water flow rate is an intermediate value between the minimum and maximum values.

Figure 4 illustrates the variation of biodiesel flow rate in function of the bottom temperature of distillation tower T-401 and water mass flow rate. The results clearly show that the bottom temperature has negligible influence over the biodiesel production. On the other hand, the water flow rate presents a maximum point of biodiesel production, although the biodiesel flow rate change was not significant

Figure 5 depicts the variation of biodiesel flow rate in relation to the reactor temperature and water mass flow rate. It is possible to observe that the reactor temperature has a great influence over the biodiesel production. The figure shows that the production grows linearly with the temperature, as expected. Furthermore, the relative change of biodiesel production regarding the water mass flow rate is insignificant if compared with the reactor temperature.





Figures 4 and 5 - Biodiesel mass flow rate vs. Distillation bottom temperature and Water mass flow rate; Biodiesel mass flow rate vs. Reactor temperature and Water mass flow rate.

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