

## EXERGY ANALYSIS OF PALM OIL BIODIESEL PRODUCTION BY BASE CATALYZED METHANOLYSIS

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**Abstract.** *In this paper the exergy analysis is applied to the process of biodiesel production by transesterification of refined palm oil with methanol using sodium hydroxide as a catalyst (base catalyzed methanolysis). The results show that the biodiesel production process has a high exergy efficiency as a consequence of the reversible nature of the transesterification reaction and the biodiesel high chemical exergy in relation to the exergy consumed in its production.*

**Keywords:** *biodiesel, palm oil, transesterification, exergy analysis*

### 1. INTRODUCTION

The global concern related to the non renewable nature of fossil fuels and the high participation of the transportation sector in the total primary energy consumption and urban pollution have become the driving forces boosting world research onto alternative engine fuels derived from biomass.

Biodiesel, defined as the mono-alkyl esters of long chain fatty acids derived from vegetable oils or animal fats, is the most widely accepted alternative fuel for diesel engines due to its technical, environmental and strategic advantages. It is biodegradable, oxygenated and practically sulfur-free. Since the carbon in the vegetable oil is originated mainly from carbon dioxide in the atmosphere, biodiesel can contribute to diminish global warming.

Biodiesel is completely miscible with petroleum diesel, thus allowing their blending in any percentage. Neat biodiesel or blended can be used in existing diesel engines without significant modifications and without an appreciable loss to engine performance.

In spite of being relatively straight-forward to produce biodiesel from several raw materials, the cost of producing this alternative fuel is greater than the conventional diesel fuel and it is often only competitive because biofuels are tax favored.

Chemically, a vegetable oil consists of triglyceride molecules (TG) which are made of three long chain fatty acids that are ester bonded to a single glycerol molecule. Biodiesel is produced by transesterification or alcoholysis of the triglycerides with short chain alcohols in the presence of a suitable catalyst. If methanol is used, the process is called methanolysis and the products of the reaction are a mixture of methyl esters (ME) and glycerol (G). The overall process is a sequence of three consecutive reversible reactions where diglycerides (DG) and monoglycerides (MG) are intermediate products.

The main variables affecting reaction kinetics and yield are the oil type and quality, the alcohol type and required excess, the type and concentration of catalyst and the reaction conditions such as temperature, pressure, reaction time and mixing intensity.

Although there are many possible options to produce it, most of the biodiesel produced today, for several reasons, is done by base catalyzed methanolysis. Methanol reacts faster than other linear and branched alcohols, (Nimcevic *et al* 2000). Homogeneous alkaline catalysts such as sodium and potassium hydroxide exhibit a good combination of price and activity, (Pinto *et al* 2005). The process proceeds at moderate pressure and temperature conditions, and yields high conversion with short reaction times, besides for reactor and vessels construction special materials are not needed.

In addition to the advantages and disadvantages of any technology for producing high standard biodiesel, strategic issues such as efficient energy utilization, environmental impacts and minimization of wastes have to be taken into account.

A primary tool to address the mentioned issues from an integral point of view is offered by exergy analysis. Exergy is defined as the maximum (theoretical) work that can be extracted from the entity (e.g. stream, amount of matter) as the entity passes from a given state to one in which it is in equilibrium with the environment.

The first attribute of exergy is that it makes it possible to compare on a common basis interactions (inputs, outputs, work, heat) that are quite different in a physical sense, (Bejan 2002). When an exergy analysis is performed on a plant

such as a chemical processing facility, the thermodynamic irreversibilities can be quantified as exergy destruction, which is wasted work or wasted potential for the production of work.

A study illustrating how the exergy analysis can be used for evaluating biodiesel production was presented by Talens *et al* (2006). The case study was the base catalyzed methanolysis of used cooking oil including an esterification pre-treatment step. The results showed that the production process has a low exergy loss which can be further minimized by using potassium hydroxide and sulphuric acid as process catalysts, and by improving the quality of the used cooking oil.

The objective of the present work is to apply the exergy analysis to evaluate the production of biodiesel from refined palm oil by base catalyzed methanolysis using sodium hydroxide as transesterification catalyst.

## 2. BIODIESEL PRODUCTION PROCESS

The data used in this study come from a pilot plant designed to test several raw materials for biodiesel production with a capacity to process 1000 kg of oil per day. Fig. 1 shows a scheme of the biodiesel production facility indicating the different steps comprising the process and identifying by a number the inputs and outputs of mass flow and energy rates.

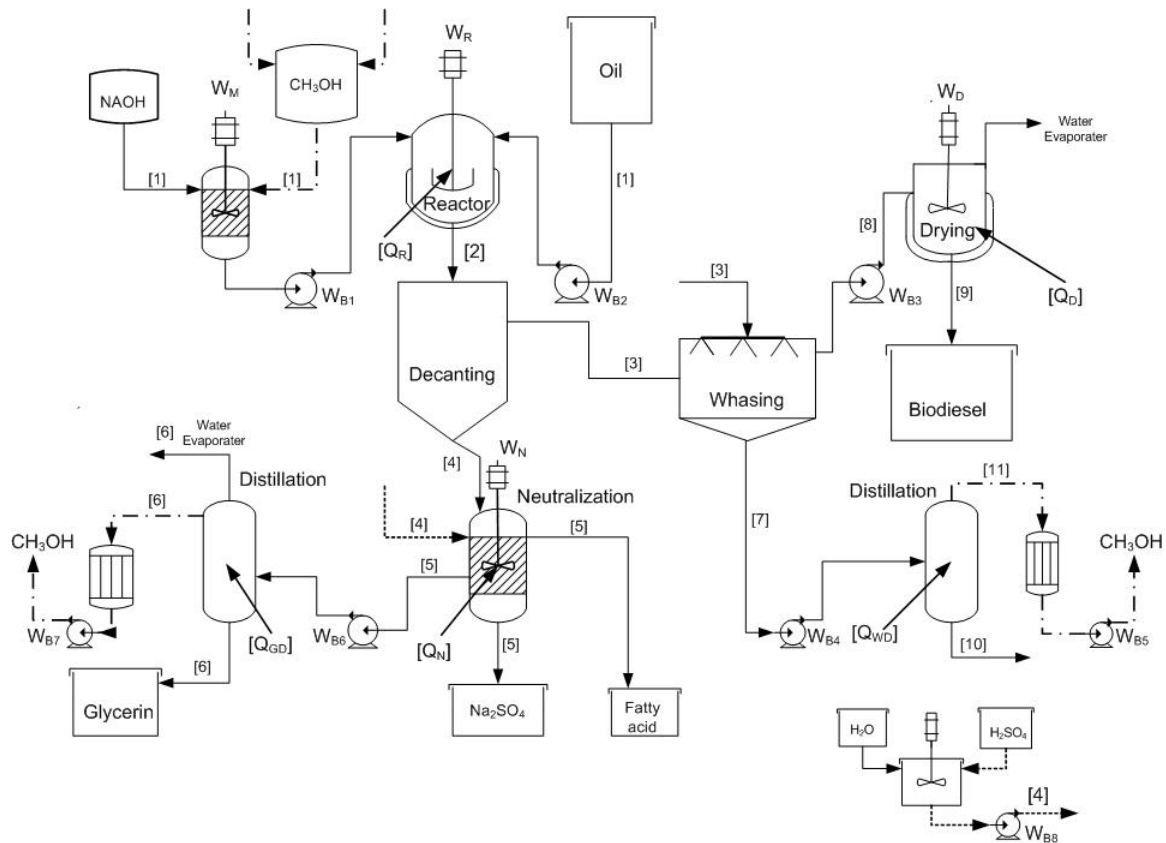


Figure 1. Scheme of the biodiesel production process

The first step is the mixing of methanol with the selected catalyst (NaOH). The reaction taking place is exothermic and leads to water formation ( $\text{CH}_3\text{-OH} + \text{NaOH} \rightarrow \text{CH}_3\text{ONa} + \text{H}_2\text{O}$ ). However, the heat released is not recovered and the quantity of water formed is low and does not affect the course of the transesterification reaction. For refined palm oil (RBD) a 6:1 molar ratio of methanol to oil (100% excess alcohol) and a 0.6% by weigh of NaOH are recommended by Darnoko and Cheryan (2000).

The second and main step is the transesterification reaction. ( $\text{TG} + 3\text{CH}_3\text{OH} \leftrightarrow 3\text{ME} + \text{G}$ ). The alcohol-catalyst mixture is combined with the palm oil in the reactor and agitated for 1 hour at 60°C. Once the reaction is complete, the reactor content is separated in two phases, one rich in ME and the other rich in glycerol.

The separation step can be promoted by gravity using a settling vessel. The lighter ME rich phase can also contain catalyst and free glycerol traces, variable concentrations of bonded glycerol (TG, DG and MG, depending on the reaction yield), soaps (proportional to the oil FFA content), and a substantial amount of the excess methanol.

On the other hand, the denser rich glycerol phase contains the major part of the catalyst used and soap formed, the rest of the excess methanol and any water formed in the occurring reactions.

After separation from the denser phase, the ME rich phase is washed gently with fresh water. In this step it is necessary to guarantee a closed contact between water and the washed phase in order to remove almost all the present methanol. This removal is favoured by the chemical affinity between water and methanol. The water also removes any soap formed.

Following the washing step, any remaining water is removed from the ME phase by a vacuum flash process or a normal distillation. Once dried, the biodiesel can be sent to storage.

On the other hand, the used water must be treated in order to be reused in the process or to be adequately disposed, and specially for recovering the methanol.

The denser phase is only about 50% glycerol and so it has little value and its disposal may be difficult. Also the methanol content requires the glycerol to be treated as hazardous waste. The glycerol refining step begins with the addition of a diluted acid, such as phosphoric or sulphuric one, to split any soap into FFA and salts. The added acid also neutralizes the base catalyst. This neutralization step required heat and mixing. The FFA are not soluble in the glycerol and will rise to the top where they can be removed. The salt precipitates out and can be filtered and dried. The methanol and water in the glycerol are removed by evaporation.

### 3. ENERGY AND EXERGY BALANCES

#### 3.1. Energy balance

The energy balance can be applied to the different steps of the biodiesel production process:

$$\sum_i \dot{Q}_i = \left( \sum_i \dot{m}_i * (h_f^0 + \Delta h) \right)_{out} - \left( \sum_i \dot{m}_i * (h_f^0 + \Delta h) \right)_{in} + \sum_i \dot{W}_i \quad (1)$$

In Equation (1),  $\dot{Q}_i$  and  $\dot{W}_i$  are the heat and work rates interacting with the control volume, the terms in the right side account for the energy in the mass, leaving and entering the control volume per unit of time,  $h_f^0$  represents the enthalpy of formation and  $\Delta h$  the enthalpy change per unit of mass, which can be calculated according to the expression:

$$\Delta h = \int_{T_{m_i}}^{T_{out}} C_p \Delta T \quad (2)$$

For pure substances such as glycerol, methanol, water, sulphuric acid, etc., the constant pressure specific heat ( $C_p$ ) can be obtained from tabulated values or data bases. On the other hand, for organic liquid mixtures such as palm oil and its biodiesel, this property must be estimated by empirical approaches such as the Lee-Keesler method, (Smith and Van Ness 2001).

$$\frac{C_p - C_p^0}{R} = (\Delta C_p)^{(0)} + \omega (\Delta C_p)^{(1)} \quad (3)$$

In equation (3)  $C_p$  and  $C_p^0$  are the constant pressure specific heats for liquids and ideal gases, respectively.  $R$  is the ideal gas constant,  $\omega$  is the acentric factor and  $(\Delta C_p)^{(0)}$  and  $(\Delta C_p)^{(1)}$  are tabulated factors.

The correlation parameters of the Lee-Keesler method are critical temperature, critical pressure, acentric factor and the ideal gas constant pressure specific heat. These parameters can be calculated using the Joback's contribution-group method, (Reid et al 2000).

$$P_{ci} = \left[ 0.113 + 0.0032 N_a - \sum_k N_k (pck) \right]^{-2} \quad (4)$$

$$T_{c,i} = T_b \left[ 0.584 + 0.965 \left( \sum_k N_k (tck) \right) - \left( \sum_k N_k (tck) \right)^2 \right]^{-1} \quad (5)$$

$$T_b = 198 + \sum_k N_k (tbk) \quad (6)$$

In Equations (4), (5) and (6),  $N_a$  is the quantity of atoms in the compound,  $N_k$  is the number of organic groups,  $pck$  is the critical pressure of each group in the compound,  $tck$  is the critical temperature of each group in the compound,  $T_b$  is the saturation temperature at normal pressure, and  $tbk$  is the saturation temperature of each group in the compound.

The constant pressure specific heat for ideal gases  $C_p^0$  is given by the following equation:

$$Cp_i^0 = \left( \sum_k N_k Cp_{ak} - 37.93 \right) + \left( \sum_k N_k Cp_{bk} + 210 \right) * T + \left( \sum_k N_k Cp_{bk} - 3.91 * 10^{-4} \right) * T^2 + \left( \sum_k N_k Cp_{bk} + 2.06 * 10^{-7} \right) * T^3 \quad (7)$$

Where  $Cp_{bk}$  is the constant pressure specific heat of each group in the compound.

The acentric factor ( $w$ ) is obtained by the following expression:

$$w = - \frac{\ln \left( \frac{P_c}{1.01325} \right) + f^{(0)}}{f^{(1)}} \quad (8)$$

Where  $f^{(0)}$  and  $f^{(1)}$  are factors given by Eq. (9) and (10):

$$f^{(0)} = \frac{-5.97616\tau + 1.29874\tau^{1.5} - 0.60394\tau^{2.5} - 1.06841\tau^5}{T_{br}} \quad (9)$$

$$f^{(1)} = \frac{-5.03365\tau + 1.11505\tau^{1.5} - 5.41217\tau^{2.5} - 7.46628\tau^5}{T_{br}} \quad (10)$$

In Equations (9) and (10),  $T_{br}$  is the ratio between the saturation temperature at normal pressure and the critical temperature and  $\tau$  is defined as following:

$$\tau = 1 - T_{br} = 1 - \frac{T_b}{T_c} \quad (11)$$

### 3.2. Exergy balance

The exergy balance for a control volume at steady-state, taking into account the concepts of physical exergy ( $b^{ph}$ ) and chemical exergy ( $b^{ch}$ ), is given by the following equation, (Kotas, 1995):

$$\left( \sum_i \dot{m}_i (b^{ch} + b^{ph})_i \right) + \dot{B}^Q = \left( \sum_j \dot{m}_j (b^{ch} + b^{ph})_j \right) + \dot{W} + \dot{I} \quad (12)$$

The left side of equation (12) represents the exergy associated to the mass entering the system ( $\dot{m}_i$ ) and to heat transfer ( $\dot{B}^Q$ ). On the other hand, the right side accounts for the mass leaving the system, the transfer work ( $\dot{W}$ ) and the quantity of which exergy is destroyed due to irreversibilities ( $\dot{I}$ ).

For a given reference environment, the values of  $b^{ch}$  for many substances are tabulated by Szargut (1988). In the case of organic compounds, which can be theoretically broken into known functional groups, chemical exergy can be estimated by means of group-contribution methods according to the following expression:

$$b_i^{ch} = \sum_k N_k * b_k^{ch} \quad (13)$$

Where  $N_k$  represents the number of the  $k$ th group in the molecule and  $b_k^{ch}$  the group chemical exergy. The chemical exergy of a mixture is a balance between the sum of the chemical exergy of substances forming the mixture and the

exergy loss due to the mixing of the substances. The chemical exergy of mixtures such as palm oil and its biodiesel when are treated as real solutions can be calculated as a function of the molar fractions ( $y_i$ ) and activity coefficients ( $\gamma_i$ ) of the individual components according to the following equation:

$$b_{mix}^{ch} = \sum_i y_i * b_i^{ch} + \bar{R} T_0 \sum_i y_i \ln(\gamma_i * y_i) \quad (14)$$

For organic compounds whose elemental compositions are known, there is an optional method for calculating their chemical exergy as a function of the lower heating value ( $LHV$ ):

$$b^{ch} = \beta * LHV \quad (15)$$

For a liquid having exclusively atoms of carbon, hydrogen and oxygen, the value of  $\beta$  can be obtained as follows:

$$\beta = 1.0374 + 0.159 \frac{H}{C} + 0.0567 \frac{O}{C} \quad (16)$$

In absence of experimental data, the  $LHV$  can be estimated, assuming complete combustion with the enthalpy of formation ( $h_f^o$ ) calculated by means of the Joback's group-contribution method, as follows:

$$h_f^o = 68.29 + \sum_k N_k hf_k \quad (17)$$

Where  $hf_k$  is the group enthalpy of formation.

The physical exergy of the mass flows rates is calculated as a function of the enthalpy and entropy changes:

$$b^{ph} = (h - h_0) - T_0 (s - s_0) \quad (18)$$

### 3.3. Performance indicators

For assessing the performance of a plant, several indicators can be defined. In this work four parameters are taken into account with this aim.

The mass performance ( $\eta_m$ ) is defined as the relation between the mass of biodiesel produced and the total mass used in the transesterification reaction:

$$\eta_m = \frac{m_B}{m_{NaOH} + m_{CH_3OH} + m_{PO}} * 100 \quad (19)$$

In this equation  $m_B$  is the mass of biodiesel produced; and  $m_{NaOH}$ ,  $m_{CH_3OH}$ , and  $m_{PO}$  are the masses of sodium hydroxide, methanol and palm oil used in the reaction.

The density of land ( $\eta_L$ ) is defined as the relation between the mass of biodiesel produced and the required land of palm oil cultivated.

$$\eta_L = \alpha * \frac{m_B}{m_{PO}} \quad (20)$$

The  $\alpha$  factor is the quantity of palm oil annually produced by hectare of land cultivated which can be assumed to be 4000 kg/ha (Jalani et al, 1997).

A parameter called as the density of energy is defined by the following equation:

$$\rho_E = \alpha * \frac{m_B * LHV}{m_{PO}} \quad (21)$$

Where LHV is the biodiesel lower heating value.

The exergy efficiency, a valuable indicator based on the concept of exergy, defined by Kotas,(1995) as rational exergy efficiency is defined as:

$$\psi = \frac{\sum \dot{B}_{out}}{\sum \dot{B}_{in}} * 100 \quad (22)$$

The terms  $\dot{B}_{out}$  and  $\dot{B}_{in}$  account for exergy outputs and inputs of the process, respectively. The rational exergy for reversible process is one. For any other case this indicator is less than one depending on the degree of irreversibility of the process.

## 4. RESULTS

### 4.1. Mass Balance

The overall mass balance, presented in Tab. 1, was obtained taking as a basis 1000 kg of processed palm oil. The numbers used to identifying the streams are the same used in Fig. 1. An important fact to stand up is the quantity of biodiesel produced with relation to the palm oil processed. For the case of study (refined palm oil) there is no formation of soaps in the transesterification reaction, and so the yield is practically 100%.

There is some lost of water associated with biodiesel drying process and methanol and glycerol purification. Almost the entire methanol used in excess is recovered.

The sodium hydroxide used as catalyser and the sulphuric acid used as neutralizer reacted to form sodium sulphate.

Table 1. Mass balance for refined palm oil (kg)

Stream	1	2	3	4	5	6	7	8	9	10	11
<b>TG</b>	1000										
<b>Methanol</b>	220	110	55	55	55	54	55				54
<b>NaOH</b>	6.0	6.0		6.0							
<b>Biodiesel</b>		1000	1000				10	990	990	10	
<b>Glycerol</b>		110		110	110	109					
<b>Water</b>			355	29.4	32.1	29.0	351	4.0		351	
<b>H<sub>2</sub>SO<sub>4</sub></b>				7.4							
<b>Na<sub>2</sub>SO<sub>4</sub></b>					10.7						

### 4.2. Properties Calculation

Table 2 presents the chemical composition of palm oil biodiesel determined by gas chromatography analysis which is used to calculate thermodynamic properties. Although palm oil is a mixture of triglycerides, its fatty acid profile must be the same of its biodiesel. In this work, it is assumed that palm oil is a mixture of four simple triglycerides: tripalmitine, tristearin, triolein and trilinolein.

Table 2. Chemical composition of palm oil biodiesel

Methyl ester type	Mass percent
Lauric acid	0.267
Miristic acid	1.434
Palmitic acid	46.130
Stearic acid	3.684
Oleic acid	37.470
Linoleic acid	11.020
Saturated methyl esters	51.515
Unsaturated methyl esters	48.485

The main thermodynamic properties of the different substances involved in the biodiesel production process are presented in Tab. 3. The constant pressure specific heat, molecular weight and enthalpy of formation for palm oil and its biodiesel were obtained from their composition, according to the methods presented in section 3.1. For the other substances these properties were obtained from tables.

The chemical exergy values for pure substances given in column 5 were obtained from tabulated values (Szargut, 2005). In the case of palm oil, biodiesel and glycerol, these values were calculated according to Eq. (13) and (14) assuming mixture ideal behavior.

Table 3. Thermodynamic properties of palm oil biodiesel and related substances

1 Substance	2 $C_p$ kJ/kgK	3 PM kg/kmol	4 $\Delta h_f^o$ kJ/mol	5 $b^{ch}$ kJ/kg	HHV (kJ/kg)		8 LHV kJ/kg	9 $\beta$	10 $b^{ch}$ kJ/kgK
					6 Theor	7 Meas			
Palm Oil	1.71	853.0	-1897.6	39573	39874	39249	37261	1.073	39990
Methanol	2.68	32.04	-238.7(l)	22408					
NaOH	1.48	40	425.6(s)	1873					
Biodiesel	1.88	285	-661.7	39842	40264	39840	37562	1.074	40211
Glycerol	2.63	92.06	-567.2	18402	19081		17169	1.14	19121
H <sub>2</sub> SO <sub>4</sub>	1.4	81.96	-814.0(l)	1666					
H <sub>2</sub> O	4.18	18	-285.8	50					
Na <sub>2</sub> SO <sub>4</sub>		142.04	-331.6	150.7					

On the other hand, the values for the same property given in column 10 were obtained using Eq. (15) and (16). As can be seen in Tab 3, the results obtained by the two mentioned routes are similar, according to the expected equations margin of error.

The higher heating values (HHV) in column 6 were obtained assuming complete combustion and using the enthalpies of formation, while the values in column 7 were determined experimentally. A comparison of such results shows the reliability of the followed approach. The lower heating values (LHV) in column 8 were obtained using the HHV given in column 6, and the elemental composition defined in Tab. 2, for palm oil and its biodiesel, and the chemical equation for glycerol.

### 4.3. Energy and exergy balances

The energy and exergy balances were carried out using the software *Engineering Equation Solver (EES)* and its library of properties for water and methanol.

For the different steps of the biodiesel production process the energy balance was carried out in order to quantify the required heat and work. The results of the balances are shown in Tab. 4. The symbols used in this table agree with those used in Fig. 1.

Table 4. Energy balance results (kJ /1000 kg of processed palm oil)

$Q_R$	77405.0	$W_{B1}$	37.0	$W_{B5}$	9.0	$W_M$	135.6
$Q_N$	15993.0	$W_{B2}$	163.5	$W_{B6}$	33.0	$W_R$	4414
$Q_D$	151934.0	$W_{B3}$	161.9	$W_{B7}$	9.0	$W_D$	3564
$Q_{WD} + Q_{GD}$	279447	$W_{B4}$	66.4	$W_{B8}$	6.0	$W_N$	726.5
$Q_T$	524779	$W_T$	9325	<b>Energy consumption total → 534,104.0</b>			

The exergy balance is carried out taking the overall plant as the control volume. The required heat is supplied using steam which delivers its enthalpy of vaporization at 3 bar. Except steam, the other streams involved in the process enter and leave the system at normal environmental conditions and so their physical exergy is zero.

The exergy balance is made using the exergy values in column six of Tab. 3, because they were obtained with the same calculate method (Szargut, 2005). The results of the exergy balance are presented in Table 5.

Table 5. Results of the exergy balance (MJ /1000 kg of processed palm oil)

	$B_{IN}$		$B_{OUT}$
NaOH	11.2	Biodiesel	39440.0
CH <sub>3</sub> OH	4930.0	CH <sub>3</sub> OH	2440.0
Oil	39570.0	Glycerol	2004.0
Water	19.2	Water	17.5
H <sub>2</sub> SO <sub>4</sub>	12.3	Na <sub>2</sub> SO <sub>4</sub>	1.6
Steam	558.0	Steam	34.8
$W_T$	9.3		
$\sum B_{IN}$	45110	$\sum B_{OUT}$	43937.9
<b>I →</b>			<b>1 172.1</b>

#### 4.4 Calculation of the Indicators

According to the mass, energy and exergy balances results, it is possible to obtain the indicators defined in section 3.3 (see Tab. 6).

Table 6. Indicators Results

Indicator	Unity	Value
Mass performance ( $\eta_m$ )	%	89.5
Agricultural performance ( $\eta_L$ )	kg/ha	3960
Energetic density ( $\rho_E$ )	MJ/ha	157.6
Exergetic efficiency ( $\psi$ )	%	92.3

For calculating the rational exergy efficiency, biodiesel, recovered methanol and water for washing were taken as outlets, in spite of glycerol and sodium sulphate being useful by-products for other process.

The rational exergy indicator shows a process with high exergy efficiency. This result can be explained by the reversibility of the transesterification reaction and the biodiesel high chemical exergy in relation to the exergy consumed in its production.

#### 5. CONCLUSIONS

The group-contribution methods constitute a valuable tool in calculating the properties required in order to carry out the exergy balance for process dealing with organic compounds.

The exergy efficiency of the base catalyzed methanolysis of palm oil is high due to the reversibility of the chemical reaction and the biodiesel high chemical exergy in relation to the exergy consumed in its production.

#### 6. ACKNOWLEDGMENTS

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