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Process Simulation and Exergy Analysis of Microalgal Biodiesel Production using *Chlorella vulgaris* via $ZnCl_2$ Pretreatment

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Abstract

Background: Exergy analysis has been recognized as a feasible approach to evaluate and improve industrial processes by identifying major irreversibilities in a system. **Objectives:** This work attempts to apply exergy analysis to a third-generation biodiesel production from *Chlorella vulgaris* microalgae. **Methods/Analysis:** Commercial industrial process simulation software was used to simulate this process. The specific exergy of many substances were found in literature and the others were calculated using Szargut, Morris & Steward's equation. A global exergy balance around the system was carried out in order to determine total irreversibilities. The contribution of unit operations and equipment to total irreversibilities was also considered. In addition, exergy efficiency and exergy emission were calculated for each stage (pretreatment, reaction, separation, biodiesel purification, and glycerol treatment). **Findings:** The global exergy efficiency was calculated in 86% similar to the results reported in other researches. The equipment that contributes the most to total irreversibility was the separation column used to remove alcohol with 487.55 kJ/kg BD. In addition, the highest irreversibilities (5.22 MJ/kg BD) and exergy emission (2.71 MJ/ kg BD) per stage were reached during biodiesel purification. **Novelty/Improvement:** The application of exergy analysis allowed to identify potential improvements in this case of study, mainly in biodiesel purification stage and process modifications are suggested to reduce total irreversibilities as reutilizing methanol and glycerol streams.

Keywords: Biodiesel, Exergy Analysis, Microalgae, Process Simulation, CAPE

1. Introduction

Growing interest is being paid to biofuels to meet the energy demand worldwide, which can be produced from different feedstocks including food crops, microalgae, agricultural residual biomass, animal fats, and cooking oils[1]. However, third-generation biofuels from microalgae are considered as a sustainable alternative approach because they do not compete with food crop and might have better environmental performance than first-generation biofuels[2]. Several kind of biofuels are potential substitutes of conventional fossil fuels, among these, biodiesel offers significant advantages related to the environment and socio-economy[3]. Biodiesel is the mono alkyl ester derived from vegetables oils, microalgae or animal tallow and is produced by means of a transesterification reaction[4,5]. Microalgae are fast growing photosynthetic organisms with fewer and more predictable process vari-

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ables (sunlight, temperature) than higher plant systems[6]. Global attempts have been made to use different species of microalgae to produce biodiesel considering the fast growth, availability, and high productivity[7]. Many fresh water microalgal species have been investigated for this purpose and *Chlorella vulgaris* is surely the most studied unicellular microorganism belonging to group *Chlorophyta* exhibiting oil content composition of 50 %[8,9]. The present study aims to apply exergy analysis to third-generation biodiesel production from *Chlorella vulgaris* in order to determine global exergy efficiency and identify process stages with high irreversibilities and exergy emissions.

2. Material and Methods

2.1 Process Description

The production of biodiesel from *Chlorella vulgaris* microalgae is the study case considered in this work. The first stage of this process is the pretreatment of free fatty acids using glycerol as shown in Figure 1. The glycerol is sent to apretreatment reactor at 200°C in presence of $ZnCl_2$ in order to produce triglycerides and water. The water content is undesirable during alkaline transesterification, hence, the outlet stream of this reactor is sent to a flash vessel. The outlet stream from the flash vessel is cooled down in a heat exchanger until 60°C and then sent to transesterification process.

The alkaline transesterification reaction stage is shown in Figure 2. This reaction takes place into a continuously stirred tank reactor (CSTR) at 60°C in presence of 1% wt. of a NaOH solution as catalyst and methanol as initiator agent. The reaction products pass through a separation stage using a decanter as shown in Figure 3. The glycerol produced in transesterification reaction is separated from biodiesel and neutralized with sulfuric acid in the treatment stage presented in Figure 4.

The biodiesel is separated from methanol in a purification stage based on a distillation column as shown in Figure 5. Then, the resulting biodiesel is washed thoroughly with hot acidulated water and sent to a decanter. In order to obtain a high purity biodiesel (96.5%), this stream is subjected to distillation process.

2.2 Exergy Analysis

The specific exergy of some substances in process streams are not available in literature, hence, it was calculated using Szargut equation[10] and standard Gibbs free energy of formation (ΔG_{fo}) as follows:

$$b_{ch} = \Delta G_{fo} + \sum_i n_i b_i \qquad \text{Equation 1}$$

The standard Gibbs free energy of formation was determined through enthalpies and entropies of pure substances at 25°C. The specific exergy of bio-substances was calculated by a semi-empirical equation (Equation 2)

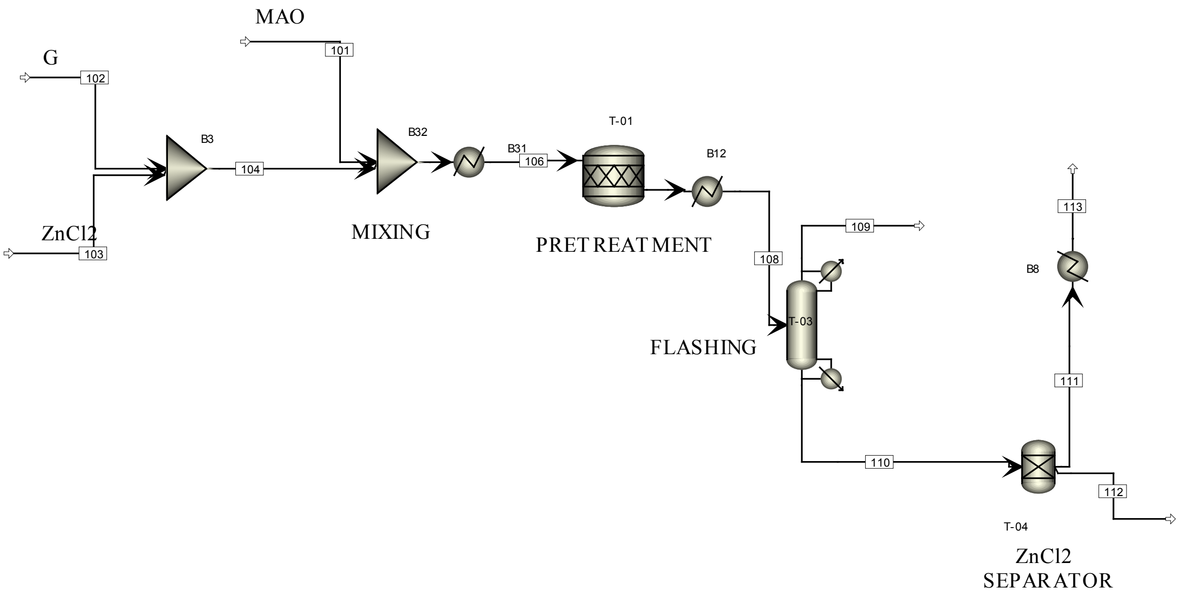

Figure 1. Flow diagram of pretreatment stage.

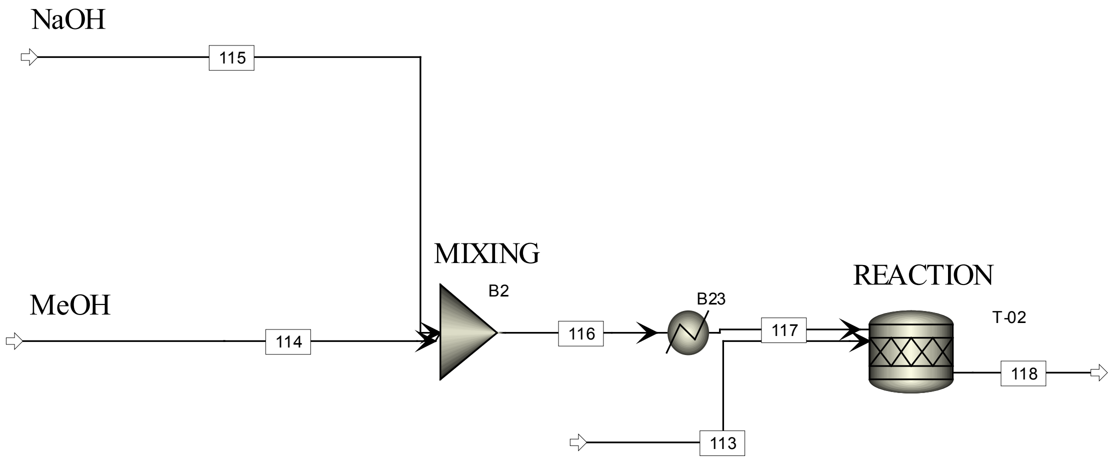

Figure 2. Flow diagram of transesterification stage.

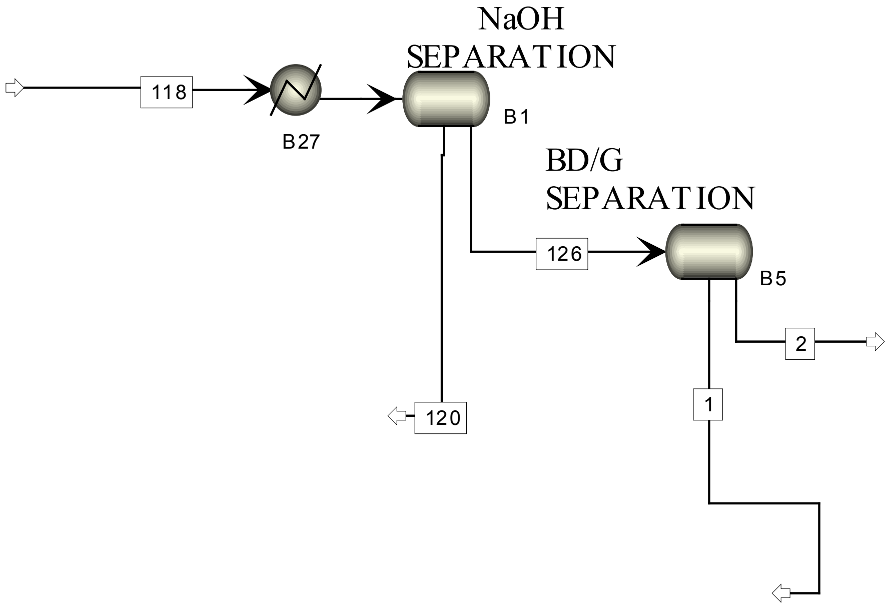

Figure 3. Flow diagram of separation stage.

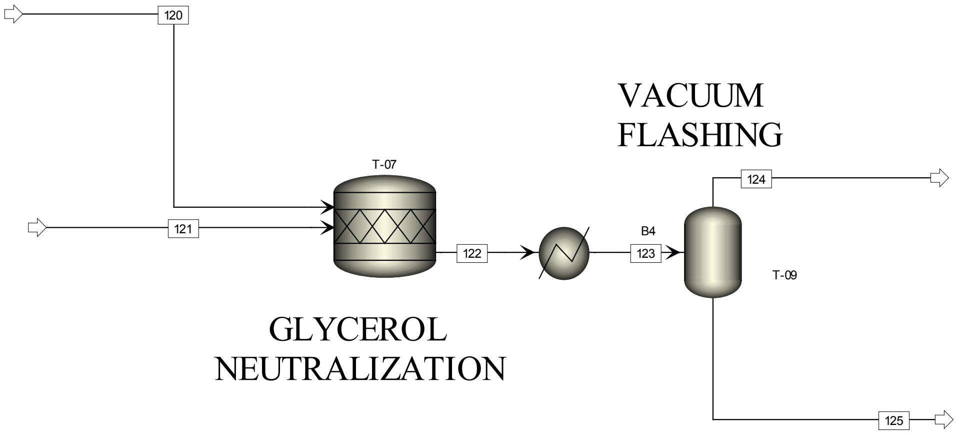

Figure 4. Flow diagram of glycerol treatment stage.

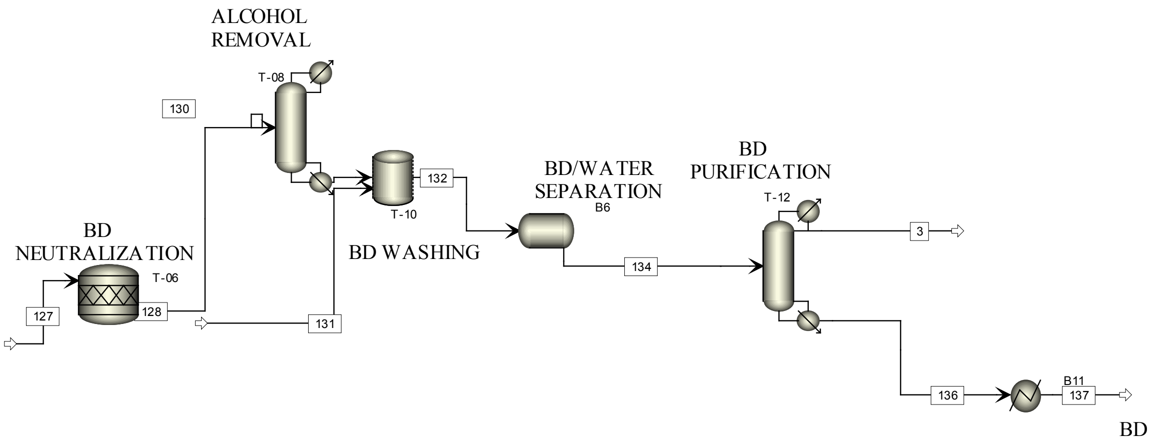

Figure 5. Flow diagram of biodiesel purification stage.

widely applied in fuels and purposed by Szargut, Morris & Steward[11], which is function of lower heating value (LHV) and elemental composition of the substance of interest.

$$b_{ch} = \beta * LHV \qquad \text{Equation 2}$$

The lower heating value was obtained by Equation 3, where higher heating value (HHV) was calculated using Equation 4 and vaporization heat of water (2442 kJ/kg). The coefficient is provided by Equation 5

$$LHV = HHV - 0.0894 * 2442.3H \qquad \text{Equation 3}$$

$$HHV = 349.1C + 1178.3H + 100{,}5S - 103.4O - 15.1N - 21.1A \qquad \text{Equation 4}$$

$$\beta = \frac{1.0438 + 0.1882\frac{H}{C} - 0.2509\left(1 + 0.7256\frac{H}{C}\right)}{1 - 0.3035\frac{O}{C}} \qquad \text{Equation 5}$$

Where C, H, S, N and A are mass composition (%) of elements and ashes in the molecules.

Exergy analysis can be used to analyse, evaluate and improve process as a measurement approach of energy quality[12]. A global exergy balance for a systems is described by Equation 6.

$$\dot{B}_{mass,in} - \dot{B}_{mass,out} + \dot{B}_{heat} - \dot{B}_{work} = \dot{B}_{loss} \qquad \text{Equation 6}$$

The total exergy of process streams are defined as a contribution of kinetic (B_K) , potential (B_P), chemical (B_{CH}) and physical exergy (B_{PH}).

$$B = B_K + B_P + B_{PH} + B_{CH} \qquad \text{Equation 7}$$

The physical exergy is given by:

$$B_{PH} = \left(H_1 - T_0S_1\right) - \left(H_0 - T_OS_0\right) \qquad \text{Equation 8}$$

Where H is the enthalpy, S is the entropy, T is the environmental temperature and subscript 1 and 0 indicates thermodynamic properties at initial and reference state, respectively.

The chemical exergy is calculated considering both chemical exergy of pure substances and mixtures.

$$B_{CH} = \sum_i N_ib_i + RT_0y_ilny_i \qquad \text{Equation 9}$$

The exergy transfer as heat is given by Equation 10.

$$B_{heat} = E_{heat}\frac{T - T_0}{T} \qquad \text{Equation 10}$$

The exergy efficiency is determined as follows:

$$\eta_B = \frac{\dot{B}_{out}}{\dot{B}_{in}} \qquad \text{Equation 11}$$

3. Results and Discussion

3.1 Process Simulation

The production of biodiesel from *Chlorella vulgaris* microalgae was simulated through commercial industrial process simulation software and operating conditions of main streams are summarized in Table 1.

It was assumed a biodiesel production rate of 7,676.37 kg/h (99%, purity) which demands 8,051.42 kg/h of microalgal biomass as feedstock. The stream of microalgal

Table 1. Operating conditions of main process streams

Stream	1	2	101	105	107	113
Flow (mol/h)	33,304.14	223.73	11,592.10	12,802.20	127,998.53	9,790.89
Temperature (°C)	25	25	25	25	200	98
Molar composition						
TG140	0.00	0.01	0.00	0.00	0.00	0.01
TG160	0.00	0.12	0.09	0.08	0.09	0.12
TG161	0.00	0.08	0.06	0.06	0.06	0.08
TG162	0.00	0.08	0.06	0.06	0.06	0.08
TG163	0.00	0.13	0.10	0.09	0.10	0.13
TG180	0.00	0.00	0.00	0.00	0.00	0.00
TG181	0.00	0.11	0.08	0.08	0.09	0.11
TG182	0.00	0.19	0.14	0.13	0.14	0.19
TG183	0.00	0.26	0.19	0.17	0.20	0.26
FAME140	0.00	0.00	0.00	0.00	0.00	0.00
FAME160	0.11	0.00	0.00	0.00	0.00	0.00
FAME161	0.07	0.00	0.00	0.00	0.00	0.00
FAME162	0.07	0.00	0.00	0.00	0.00	0.00
FAME163	0.11	0.00	0.00	0.00	0.00	0.00
FAME180	0.00	0.00	0.00	0.00	0.00	0.00
FAME181	0.10	0.00	0.00	0.00	0.00	0.00
FAME182	0.16	0.00	0.00	0.00	0.00	0.00
FAME183	0.22	0.00	0.00	0.00	0.00	0.00
FFA140	0.00	0.00	0.00	0.00	0.00	0.00
FFA160	0.00	0.00	0.03	0.03	0.00	0.00
FFA161	0.00	0.00	0.02	0.02	0.00	0.00
FFA162	0.00	0.00	0.02	0.02	0.00	0.00
FFA163	0.00	0.00	0.03	0.03	0.00	0.00
FFA180	0.00	0.00	0.00	0.00	0.00	0.00
FFA181	0.00	0.00	0.03	0.03	0.00	0.00
FFA182	0.00	0.00	0.05	0.04	0.00	0.00
FFA183	0.00	0.00	0.07	0.06	0.00	0.00
METHANOL	0.15	0.01	0.00	0.00	0.00	0.00
GLYCEROL	0.00	0.00	0.00	0.09	0.02	0.02
NA_2SO_4	0.00	0.00	0.00	0.00	0.00	0.00
WATER	0.00	0.00	0.00	0.00	0.23	0.00
H_2SO_4	0.00	0.00	0.00	0.00	0.00	0.00
NAOH	0.00	0.00	0.00	0.00	0.00	0.00
$ZNCL_2$	0.00	0.00	0.00	0.00	0.00	0.00

Table 1. Operating conditions of main process streams (*continuation*)

Stream	124	125	126	130	134	137
Flow (mol/h)	16,604.33	12,705.16	33,527.96	27,189.91	27,633.43	27,206.17
Temperature (°C)	95	95	25	388.36	25	25
Molar composition						
TG140	0.00	0.00	0.00	0.00	0.00	0.00
TG160	0.00	0.00	0.00	0.00	0.00	0.00
TG161	0.00	0.00	0.00	0.00	0.00	0.00
TG162	0.00	0.00	0.00	0.00	0.00	0.00
TG163	0.00	0.00	0.00	0.00	0.00	0.00
TG180	0.00	0.00	0.00	0.00	0.00	0.00
TG181	0.00	0.00	0.00	0.00	0.00	0.00
TG182	0.00	0.00	0.00	0.00	0.00	0.00
TG183	0.00	0.00	0.00	0.00	0.00	0.00
FAME140	0.00	0.00	0.00	0.00	0.00	0.00
FAME160	0.00	0.00	0.11	0.13	0.13	0.13
FAME161	0.00	0.00	0.07	0.09	0.09	0.09
FAME162	0.00	0.00	0.07	0.09	0.09	0.09
FAME163	0.00	0.00	0.11	0.13	0.13	0.13
FAME180	0.00	0.00	0.00	0.00	0.00	0.00
FAME181	0.00	0.00	0.10	0.09	0.09	0.09
FAME182	0.00	0.00	0.16	0.20	0.19	0.20
FAME183	0.00	0.00	0.22	0.27	0.27	0.27
FFA140	0.00	0.00	0.00	0.00	0.00	0.00
FFA160	0.00	0.00	0.00	0.00	0.00	0.00
FFA161	0.00	0.00	0.00	0.00	0.00	0.00
FFA162	0.00	0.00	0.00	0.00	0.00	0.00
FFA163	0.00	0.00	0.00	0.00	0.00	0.00
FFA180	0.00	0.00	0.00	0.00	0.00	0.00
FFA181	0.00	0.00	0.00	0.00	0.00	0.00
FFA182	0.00	0.00	0.00	0.00	0.00	0.00
FFA183	0.00	0.00	0.00	0.00	0.00	0.00
METHANOL	0.99	0.20	0.15	0.00	0.00	0.00
GLYCEROL	0.00	0.75	0.00	0.00	0.00	0.00
NA_2SO_4	0.00	0.02	0.00	0.00	0.00	0.00
WATER	0.01	0.03	0.00	0.00	0.02	0.00
H_2SO_4	0.00	0.00	0.00	0.00	0.00	0.00
NAOH	0.00	0.00	0.00	0.00	0.00	0.00
$ZNCL_2$	0.00	0.00	0.00	0.00	0.00	0.00

oil was defined in Table 2 according to characterizations reported by[13] and assuming a composition of free fatty acids of 10%[14].

3.2 Exergy Analysis

The specific exergy can be found in literature for given substances; however, complex molecules as triglycerides, fatty acids, and esters of fatty acids require to be calculated by Equations 2-5. Table 3 lists specific exergy of substances that are in process streams considering both constituent atoms and bonds energy.

Szargut, Morris & Steward's equation has been widely used for similar substances. The standard Gibbs free energy of formation was provided by the simulation software. Table 4 summarizes the comparative results obtained using both equations for fatty acids (FFA14:0), triglycerides and methyl-ester.

The relative error of Szargut, Morris & Steward's equation is not higher than 4 %. In addition, it is often recommended to use semi-empirical equation for biomass due to the specific exergy is easily determined through a coefficient (β). Liquid and gaseous fuels have exergy values similar to its lower heating value and the empirical coefficient is approximated to 1.

Chemical and physical exergies of process streams were calculated to perform exergy analysis in the case of study. The temperature of inlet streams is the surronding temperature, hence, physical exergy is zero due to the equilibrium between environment and streams. Table 5 presents total exergy of main inlet and outlet process streams.

Table 2. Composition of fatty acids of Chlorella vulgaris microalgae[13]

TG	**14:0**	**16:0**	**16:1**	**16:2**	**16:3**	**18:0**	**18:1**	**18:2**	**18:3**
Composition (%, dry basis)	0.5	12	8	8	12	0.5	12	12	27

Table 3. Specific exergies of substances in process streams

Substance	**b_{Ch} [kJ/mol]**	**Compound**	**b_{Ch} [kJ/mol]**
TG14:0	27,507.7	FFA14:0	8,668.1
TG16:0	31,333.6	FFA16:0	9,943.3
TG16:1	30,720.0	FFA16:1	9,738.7
TG16:2	30,106.7	FFA16:2	9,534.1
TG16:3	29,493.5	FFA16:3	9,329.6
TG18:0	35,159.1	FFA18:0	11,218.4
TG18:1	34,546.1	FFA18:1	11,013.9
TG18:2	33,933.2	FFA18:2	10,809.6
TG18:3	33,320.5	FFA18:3	10,605.2
FAME14:0	9,305.8	CH_3OH^*	720
FAME16:0	10,580.9	$ZnCl_2^*$	93.4
FAME16:1	10,376.3	$Na_2SO_4^*$	21.4
FAME16:2	10,171.8	$H_2SO_4^*$	109
FAME16:3	9,967.4	$NaOH^*$	77.4
FAME18:0	11,855.9	H_2O^*	0.9005
FAME18:1	11,651.6	H_2O (Steam)*	9.49127
FAME18:2	11,447.3	Glycerol*	1,725.83
FAME18:3	11,242.9		

Table 4. Comparative results for specific exergies of substances

Substances	b_{Ch} [kJ/mol]		Relative error [%]
	Szargut, Morris & Steward (1988)	Szargut (2005)	
TG14:0	27,507.69	28,015.66	1.85
TG16:0	31,333.56	31,946.51	1.96
TG16:1	30,720.01	31,477.57	2.47

Table 5. Total exergy of main inlet and outlet process streams

Streams		**Total exergy [MJ/kg BD]**
	Microalgal oil	40.17
Inlet	Glycerol	0.27
	$ZnCl_2$	9.55×10^{-5}
	Methanol	4.89
	Total	45.49
	Glycerol	2.38
Outlet	Biodiesel	38.84
	Total	43.31

The global exergy balance of biodiesel production process is shown in Figure 6. It was found that the highest inlet and outlet exergies are due to microalgal oil/glycerol and biodiesel/methanol, respectively. The usable streams were biodiesel and glycerol because of they are sources of useful energy. The others streams were considered as exergetic emissions. The glycerol was assumed as byproduct; however, it could be recirculated to pretreatment stage in order to reduce biodiesel production costs.

The total exergetic emissions were calculated in 5,597.26 kJ/kg BD, which represents 11.45% of total inlet exergy. The irreversibilities of this process was 4.83 % of total inlet exergy indicating the need of modifying the case of study to improve biodiesel production from an exergetic point of view.

Local exergy balance to main equipment was carried out in order to determine their irreversibilities and these results are listed in Table 6. It is well known that exergy is lost partial or totally due to irreversibilities inherent to process energetic transformations. For electric equipment, it was considered that 1 kW of inlet power represents 1 kW of exergy as reported by[15]. One of the

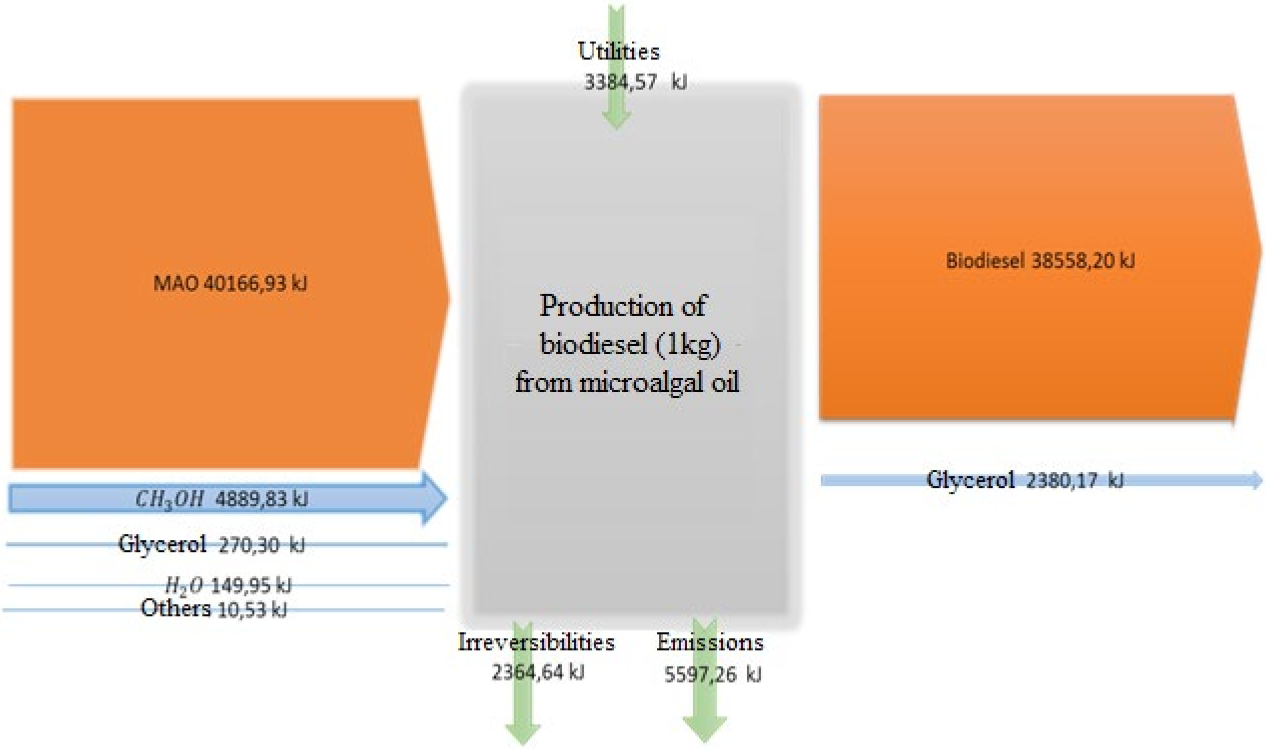

Figure 6. Global exergy balance for biodiesel production from microalgae.

Table 6. Irreversibilities per equipment in biodiesel production from microalgae

Equipment	Irreversibilities [kJ/kg BD]
Pretreatment reactor (T-01)	37.61
Transesterification reactor (T-02)	42.28
Flash evaporator (T-03)	1.39
Biodiesel neutralization reactor (T-06)	0
Glycerol neutralization reactor (T-07)	10.24
Alcohol purification column (T-08)	487.55
Vacuum flash evaporator (T-09)	7.26
Biodiesel purification column (T-12)	422.71

separation equipment in pretreatment stage (T-04) was simulated as splitter, which tends to ideality and irreversibilities were assumed of zero. The highest irreversibilities were observed for alcohol purification column (T-08) followed by biodiesel purification column (T-12) with values of 487.55 and 433.71 kJ/kg BD, respectively.

As shown in Figure 7, the unit operation of distillation exhibited the highest contributed (38.86%) to total irreversibilities, followed by decanting with 37.28%, which is attributed to destruction of exergy associated to heat transfer in reboiler and cooler located in the distillation column. Mixing and absorption operations contributed to 0.1% and 10% of total irreversibilities, respectively, because of the high entropy generation.

The irreversibilities per stage are listed in Table 7, in which biodiesel purification exhibited the highest value (5.22 MJ/kg BD). This result was expected due to the equipment used in this stage (distillation column, separator and absorption column) showed high contributions to total irreversibilities. The reaction stage achieved the lowest irreversibilities attributed to total conversion in transesterification reactor.

Generally, energetic efficiency is applied as parameter to evaluate thermodynamic performance of a system, however, exergetic efficiency offers information concerning quality of energy. Figure 8 shows exergetic efficiency of process stages (pretreatment, reaction, separation, biodiesel purification, and glycerol treatment). It was observed that the highest efficiencies were reached in pretreatment and reaction stages due to inconsiderable exergy emissions. The lowest efficiency (10.67 %) was obtained for separation stage, which can be attributed to the high amount of exergy used in decanting equipment.

The global exergetic efficiency was calculated in 86% considering the reusability of glycerol stream (stream

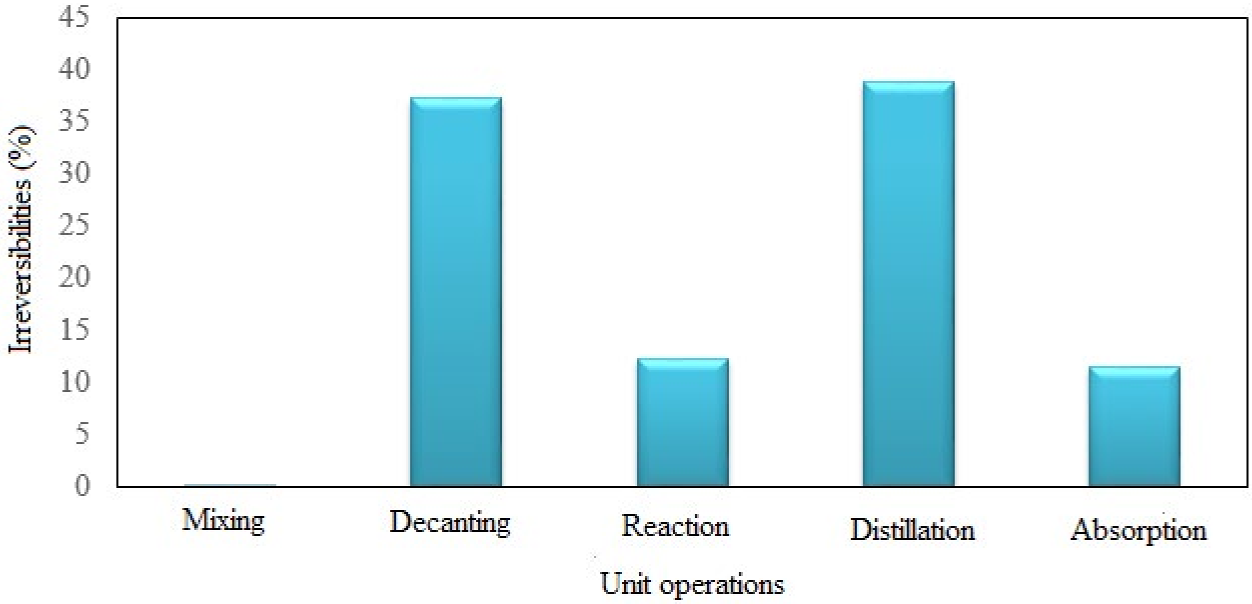

Figure 7. Contribution of unit operations to total irreversibilities.

Table 7. Irreversibilities per process stages

Stage	**Irreversibilities [MJ/kg BD]**
Pretreatment	0.199
Reaction	0.050
Separation	0.115
Glycerol treatment	0.109
Biodiesel purification	5.520

125). Other exergetic efficiencies for biodiesel production process are reported in Table 8. The highest exergetic efficiency (98%) was reached by[15], followed by this case of study.

The biodiesel purification stage exhibited the highest exergy emission (2.71 MJ/kg BD) as is shown in Figure 9, which causes a reduction in exergetic efficiency. The glycerol treatment and separation stages obtain exergy emission of 1.54 and 0.93 MJ/kg BD, respectively. It was identified different alternative for improving these results as using thermal streams to reduce industrial utilities, recirculating stream 124 (contains methanol) to transesterification reactor and reusing ZnCl$_2$ in pretreatment stage.

Table 8. Summary of exergy efficiency for biodiesel production from different feedstocks

Feedstock	**Exergy efficiency**	**Reference**
Cooking oil	98%	[15]
Jatropha curcas	44%	[16]
Chlorellasp	79%	[17]
Jatrophacurcas	63%	[18]
Brassicanapus L.	66%	[19]
Chlorella vulgaris	86%	[15]

4. Conclusions

Computer-aided exergy analysis was applied in this work to third-generation biodiesel production using *Chlorella vulgaris* microalgae in order to identify stages that most contribute to total irreversibilities. The results showed a higher value for global exergetic efficiency (86%) in comparison to other studies that usedifferent feedstocks including *Jatropha curcas* and *Brassica napus L*. The unit operation of distillation exhibited the highest contribution to total irreversibilities (38.86%) followed by decanting. In addition, the equipment with

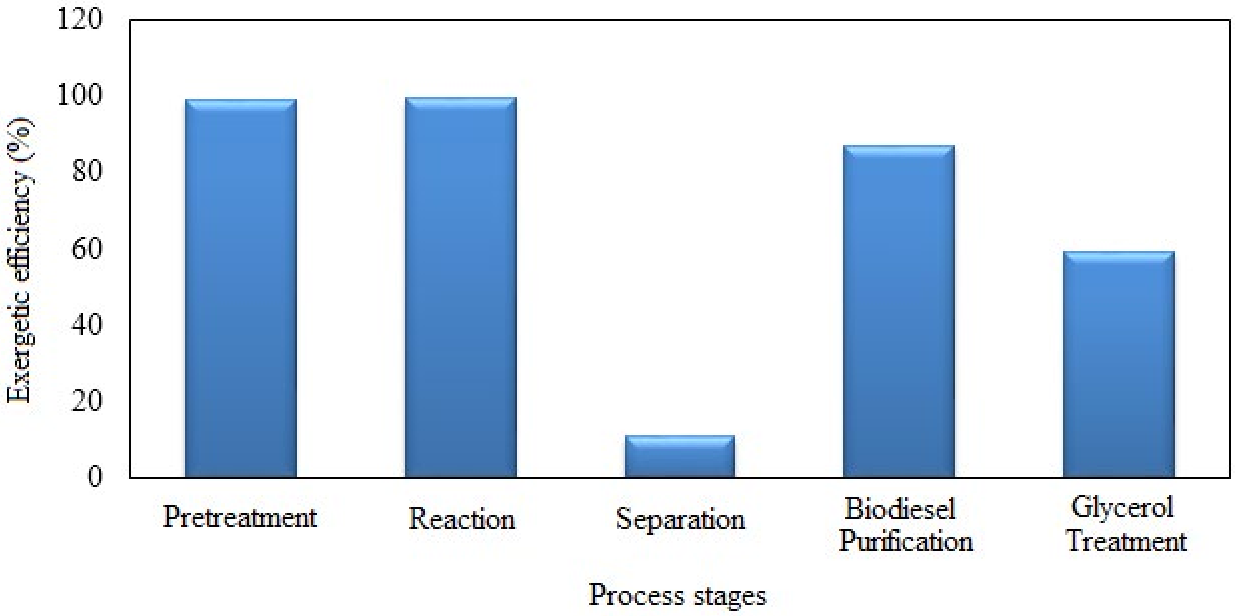

Figure 8. Exergetic efficiency of process stages.

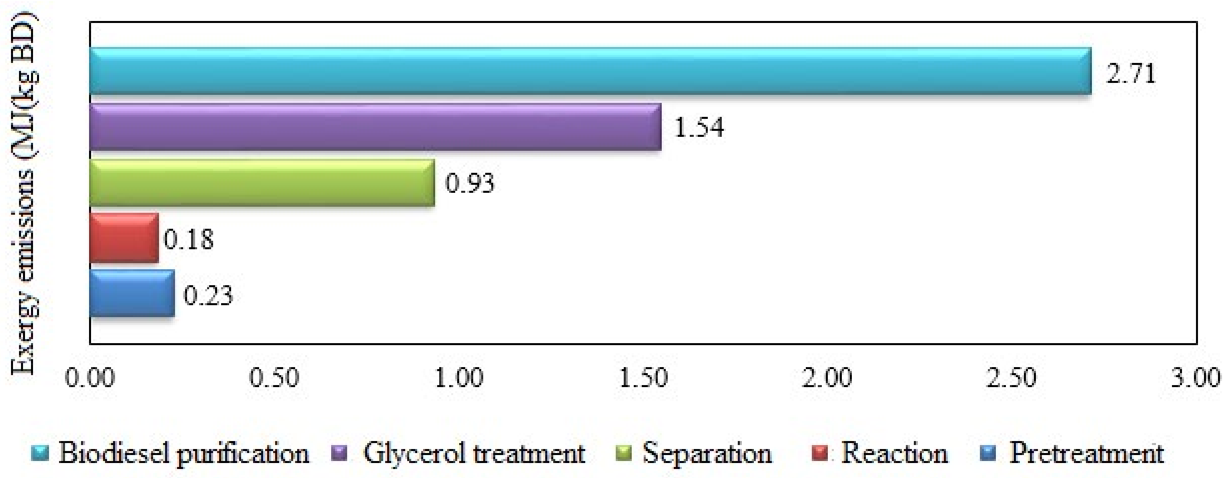

Figure 9. Exergy emission in process stages.

highest irreversibilities value (487.55 kJ/kg BD) was the separation column to remove alcohol from biodiesel. The exergy balance per stage reported that the highest irreversibilities (5.22 MJ/kg BD) and exergy emission (2.71 MJ/kg BD) were achievedduring biodiesel purification. Reutilization of methanol and glycerol are suggested in order to reduce total irreversibilities. These results reveal that exergy analysis is a useful tool to identify potential improvements to increase global exergetic efficiency.

5. Acknowledgments

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