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# MATHEMATICAL MODELING OF BATCH DISTILLATION OF HYDROCARBONS PRODUCED BY MICROALGAE

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**Abstract.** *With the burning of fossil fuels, new energy sources end up being required to prevent the increase of CO<sub>2</sub> and consequently global warming. Microalgae come as an option because they produce biodiesel, which is a source of non-toxic, biodegradable fuel that releases cleaner gases to the environment. In addition, microalgae have the potential to produce hydrocarbons, which are similar to hydrocarbons of fossil origin. However, in order to make microalgae hydrocarbons an usable fuel, a purification process is necessary since the hydrocarbons when extracted are contaminated with microalgae pigments and lipids. One type of purification is the fractional distillation, however this process consumes a lot of energy and therefore there is a need for optimization, which can be done by mathematical modeling. Therefore, the objective of this work is the mathematical modeling of batch distillation hydrocarbons produced by microalgae. The methodology used was mass balance and energy by the volume elements method, which is based on the optimization of systems engineering of physical size of study. The advantage of this methodology is that consists of a simple sequence of validation of the model with necessary adjustments until the mathematical modeling reaches a value obtained experimentally. The simulations of the model describe the system well, correctly separating the proposed compounds at the beginning of the model. The model can be used for optimization of distillation after experimental validation.*

**Keywords:** *Microalgae, hydrocarbon, distillation, mathematical model.*

## 1. INTRODUCTION

The rapid depletion of oil reserves, climate change and pollution of the environment is briskly increasing due to the growing energy demand. One way to overcome these problems are the alternative energy sources, i.e. biofuels (Chisti, 2007). This renewable energy source has garnered much attention in recent years due to a number of environmental, economic and social benefits and is emerging as an ideal substitute for fossil fuels (Demirbas, 2011).

Microalgae appear as a strong candidate in the production of biofuels, since it has the capacity to store a large quantity of lipids, reaching up to 75% of its dry biomass (Chisti, 2007). Biofuels produced by microalgae are biodegradable, renewable and non-toxic and when compared to vegetable oils have a higher growth rate and can be grown in brackish waters that are unsuitable for the growth of other crops (Alcaine, 2010; Puppen, 2002; Sheehan *et al.*, 1998). Microalgae biomass can also be used to produce bioethanol, biohydrogen, biogas and synthesis gas (Wegeberg, 2010).

In addition to lipids (long chain fatty acids), microalgae can produce long chain hydrocarbons that can be used directly as fuels or as additives. These hydrocarbons provide a biofuel that can be converted to gasoline by hydrocracking or can be mixed with diesel and jet fuel (Hillen *et al.*, 1982). The microalgae hydrocarbons may range from C<sub>12</sub> to C<sub>32</sub> (Dayan *et al.*, 2010). In order to separate these hydrocarbons contained in the microalgae, fractional distillation is essential since it is widely employed by the petrochemical industry to separate several petroleum hydrocarbon fractions (Seader *et al.*, 1998).

## 2. VOLUME ELEMENT MODEL (VEM)

The method was initially proposed for the thermal management of the electronics conditioning system, being generalized to allow its use in diverse problems that include, besides thermal analysis, other phenomena such as mass and species transport among the volume elements (EV). The governing equations are the principles of conservation of mass, energy and species applied to each EV. (Vargas, JVC. *et al.*, 2001); the system is divided into four control volumes as shown in figure 1.

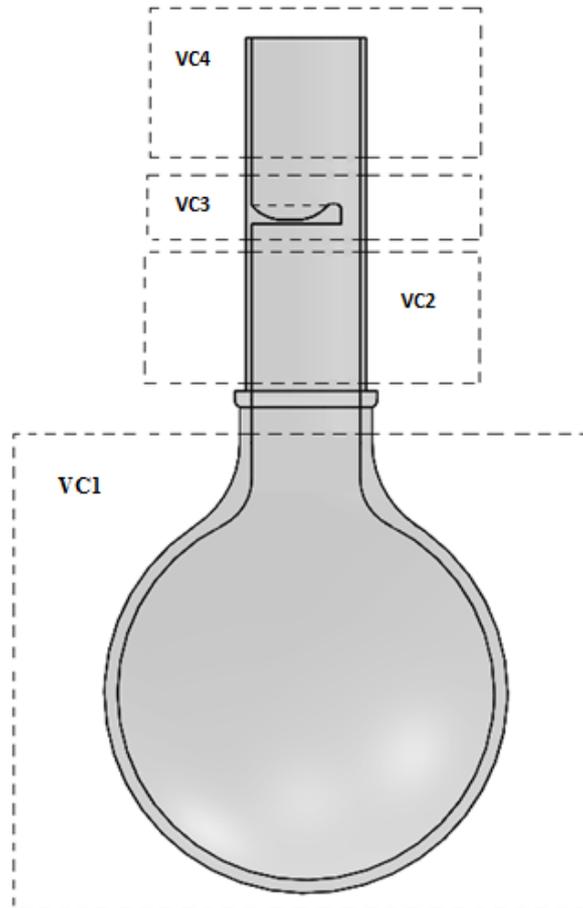


Figure 1. Volume controls. Source: The Authors (2017).

Considerations:

- Ideal gas.
- Joback method to calculate specific heats at constant pressure (Cp).
- Antoine equation.
- Only two compounds (dodecane and palmitic acid).
- Constant pressure.
- Plate with 1% mass (practically empty).

### 3. MATHEMATICAL EQUATIONS

Calculation of the flows in the balloon:

$$X_{1m} = \frac{M_1}{\rho} \quad (1)$$

$$X_{2m} = \frac{M_2}{\rho} \quad (2)$$

$$n_{total1} = \left( \frac{X_{1m}}{m_{dode}} \right) + \left( \frac{X_{2m}}{m_{acid}} \right) \quad (3)$$

$$X_{1molar} = \frac{X_{1m}}{(n_{total1} \times m_{dode})} \quad (4)$$

$$X_{2molar} = \frac{X_{2m}}{(n_{total1} \times m_{acid})} \quad (5)$$

Using the Antoine equation:

$$\log_{10} p = A - \frac{B}{C+T} \quad (6)$$

$$\log_{10} p_{1dode} = A_{dode} - \left( \frac{B_{dode}}{C_{dode} + T_1} \right) \quad (7)$$

$$P_{sat_{1dode}} = 10^{P_{1dode}} \quad (8)$$

$$C_{sat_{1dode}} = \frac{M_3}{P_{sat_{1dode}}} \quad (9)$$

$$\log_{10} p_{1acid} = A_{acid} - \left( \frac{B_{acid}}{C_{acid} + T_1} \right) \quad (10)$$

$$P_{sat_{1acid}} = 10^{P_{1acid}} \quad (11)$$

$$C_{sat_{1acid}} = \frac{M_4}{P_{sat_{1acid}}} \quad (12)$$

$$\dot{M}_{1dode} = kla_{dode} \times (X_{1molar} - C_{sat_{1dode}}) \times n_{total1} \times m_{dode} \times M_5 \quad (13)$$

$$\dot{M}_{1acid} = kla_{acid} \times (X_{2molar} - C_{sat_{1acid}}) \times n_{total1} \times m_{acid} \times M_5 \quad (14)$$

Calculation of flow rates in the column plate:

$$X_{3m} = \frac{M_6}{\rho} \quad (15)$$

$$X_{4m} = \frac{M_7}{\rho} \quad (16)$$

$$n_{total2} = \left( \frac{X_{3m}}{m_{dode}} \right) + \left( \frac{X_{4m}}{m_{acid}} \right) \quad (17)$$

$$X_{3molar} = \frac{X_{3m}}{(n_{total2} \times m_{dode})} \quad (18)$$

$$X_{4molar} = \frac{X_{4m}}{(n_{total2} \times m_{acid})} \quad (19)$$

Using the Antoine equation:

$$\log_{10} p = A - \frac{B}{C+T} \quad (20)$$

$$\log_{10} p_{2dode} = A_{dode} - \left( \frac{B_{dode}}{C_{dode} + T_2} \right) \quad (21)$$

$$P_{sat_{2dode}} = 10^{P_{2dode}} \quad (22)$$

$$C_{sat_{2dode}} = \frac{M_8}{P_{sat_{2dode}}} \quad (23)$$

$$\log_{10} p_{2acid} = A_{acid} - \left( \frac{B_{acid}}{C_{acid} + T_2} \right) \quad (24)$$

$$P_{sat_{2acid}} = 10^{P_{2acid}} \quad (25)$$

$$C_{sat_{2acid}} = \frac{M_9}{P_{sat_{2acid}}} \quad (26)$$

$$\dot{M}_{dode} = kla_{dodeplate} \times (X_{3molar} - C_{sat_{2dode}}) \times n_{total2} \times m_{dode} \times \rho \times V_{plate} \quad (27)$$

$$\dot{M}_{2acid} = kla_{acidplate} \times (X_{4molar} - C_{sat_{2acid}}) \times n_{total2} \times m_{acid} \times \rho \times V_{plate} \quad (28)$$

$$M_l = \dot{M}_{dode} + \dot{M}_{2acid} \quad (29)$$

$$M_{plate} = (M_6 + M_7) \times V_{plate} \quad (30)$$

### 3.1 Volume element 1, on the bottom:

Mass balance in element 1:

$$\frac{dM}{dt} = -\dot{M}_{dode} - \dot{M}_{acid} - M_l \quad (31)$$

Using the Joback method to determine the specific heats (Cp):

$$C_p = \sum a_i - 37.93 + [\sum b_i + 0.210] \times T + [\sum c_i - 3.91 \times 10^{-4}] \times T^2 + [\sum d_i + 2.06 \times 10^{-7}] \times T^3 \quad (32)$$

$$C_{pdode} = \frac{(A_{cpdode} + B_{cpdode} \times T_1 + C_{cpdode} \times T_1^2 + D_{cpdode} \times T_1^3)}{m_{dode}} \quad (33)$$

$$C_{pacid} = \frac{(A_{cpacid} + B_{cpacid} \times T_1 + C_{cpacid} \times T_1^2 + D_{cpacid} \times T_1^3)}{m_{acid}} \quad (34)$$

$$C_1 = (-\dot{M}_{dode} \times C_{pdode} \times T_1) - (\dot{M}_{acid} \times C_{pacid} \times T_1) + (-M_l \times M_6 \times C_{pdode} \times T_1) + (-M_l \times M_7 \times C_{pacid} \times T_1) \quad (35)$$

Energy balance in element 1:

$$\frac{dT_1}{dt} = \frac{C_1 + Q_{res} - (\frac{dM_1}{dt} \times T_1 \times \rho \times C_{pdode}) - (\frac{dM_2}{dt} \times T_1 \times \rho \times C_{pacid})}{(\rho \times C_{pdode} \times M_1) + (\rho \times C_{pacid} \times M_2)} \quad (36)$$

Dodecane mass balance:

$$\frac{dM_1}{dt} = \frac{(-\dot{M}_{dode} \times \rho) + (-M_l \times M_6 \times \rho) - (M_1 \times \frac{dM_5}{dt})}{M_5} \quad (37)$$

Mass balance of palmitic acid:

$$\frac{dM_2}{dt} = \frac{(-\dot{M}_{acid} \times \rho) + (-M_l \times M_7 \times \rho) - (M_2 \times \frac{dM_5}{dt})}{M_5} \quad (38)$$

### 3.2 Volume element 2, gas above the balloon:

Total gas flow:

$$Xm_{total} = \left( \frac{\dot{M}_{dode}}{m_{dode}} \right) + \left( \frac{\dot{M}_{acid}}{m_{acid}} \right) \quad (39)$$

Dodecane mass balance:

$$\frac{dM_3}{dt} = \left( \frac{\dot{M}_{dode}}{m_{dode}} - \frac{Xm_{total} \times M_3}{P_{total}} \right) \times \frac{R_{gás} \times M_{10}}{V_{gás}} + \frac{M_3 \times \frac{dT_3}{dt}}{\frac{dT_3}{dt}} \quad (40)$$

Mass balance of palmitic acid:

$$\frac{dM_4}{dt} = \left( \frac{\dot{M}_{acid}}{m_{acid}} - \frac{Xm_{total} \times M_4}{P_{total}} \right) \times \frac{R_{gás} \times M_{10}}{V_{gás}} + \frac{M_4 \times \frac{dT_3}{dt}}{\frac{dT_3}{dt}} \quad (41)$$

$$C_{p2dode} = \frac{(A_{cpdode} + B_{cpdode} \times T_3) + (C_{cpdode} \times T_3^2 + D_{cpdode} \times T_3^3)}{m_{dode}} \quad (42)$$

$$C_{p2acid} = \frac{(A_{cpacid} + B_{cpacid} \times T_3) + (C_{cpacid} \times T_3^2 + D_{cpacid} \times T_3^3)}{m_{acid}} \quad (43)$$

$$M_{1gás} = \frac{M_3 \times V_{gás} \times m_{dode}}{R_{gás} \times T_2} \quad (44)$$

$$M_{2gás} = \frac{M_4 \times V_{gás} \times m_{acid}}{R_{gás} \times T_2} \quad (45)$$

$$C_2 = (M_{1gás} \times C_{p2dode}) + (M_{2gás} \times C_{p2acid}) \quad (46)$$

$$P_3 = P_{total} - M_3 - M_4 \quad (47)$$

Energy balance in gas:

$$\frac{dT_3}{dt} = \frac{(\dot{M}_{dode} \times C_{pdode} \times T_1) + (\dot{M}_{acid} \times C_{pacid} \times T_1) - (Xm_{total} \times m_{dode} \times M_3) + \frac{(P_3 \times C_{p2dode} \times T_3)}{P_{total}} - (Xm_{total} \times m_{acid} \times M_4) \times \frac{(C_{p2acid} \times T_3)}{P_{total}}}{C_2} \quad (48)$$

### 3.3 Volume element 3, liquid in plate:

Dodecane mass balance:

$$\frac{dM_6}{dt} = \frac{(-\dot{M}_{2dode} + M_l \times M_6)}{V_{plate}} \quad (49)$$

Mass balance of palmitic acid:

$$\frac{dM_7}{dt} = \frac{(-\dot{M}_{2acid} + M_l \times M_7)}{V_{plate}} \quad (50)$$

$$C_{p3dode} = \frac{(A_{cpdode} + B_{cpdode} \times T_2) + (C_{cpdode} \times T_2^2 + D_{cpdode} \times T_2^3)}{m_{dode}} \quad (51)$$

$$C_{p3acid} = \frac{(A_{cpacid} + B_{cpacid} \times T_2) + (C_{cpacid} \times T_2^2 + D_{cpacid} \times T_2^3)}{m_{acid}} \quad (52)$$

$$C_{p4dode} = \frac{(A_{cpdode} + B_{cpdode} \times T_4) + (C_{cpdode} \times T_4^2 + D_{cpdode} \times T_4^3)}{m_{dode}} \quad (53)$$

$$C_{p4acid} = \frac{(A_{cpacid} + B_{cpacid} \times T_4) + (C_{cpacid} \times T_4^2 + D_{cpacid} \times T_4^3)}{m_{acid}} \quad (54)$$

$$C_3 = (-\dot{M}_{2dode} \times C_{p4dode} \times T_4) - (\dot{M}_{2acid} \times C_{p4acid} \times T_4) + (M_l \times M_6 \times C_{p3dode} \times T_2) + (M_l \times M_7 \times C_{p3acid} \times T_2) \quad (55)$$

Energy balance in the plate:

$$\frac{dT_2}{dt} = \left( \frac{C_3}{V_{plate} \times C_{p3dode} + C_{p3acid}} \right) - \frac{(T_3 \times M_6) - (T_3 \times M_7)}{(M_6 + M_7)} \quad (56)$$

### 3.4 Volume element 4, gas above the plate:

$$Xm_{2total} = \left( \frac{Xm_{total} + \dot{M}_{2dode}}{m_{dode}} \right) + \left( \frac{\dot{M}_{2acid}}{m_{acid}} \right) \quad (57)$$

Dodecane mass balance:

$$\frac{dM_8}{dt} = \left(\frac{\dot{M}_{dode}}{m_{dode}}\right) + \left(\frac{Xm_{total} \times M_3}{P_{total}}\right) - \left(\frac{Xm_{2total} \times M_8}{P_{total}}\right) \times \left(\frac{R_{gás} \times T_4}{V_{gás}}\right) + \left(\frac{M_8 \times \frac{dT_4}{dt}}{T_4}\right) \quad (58)$$

Mass balance of palmitic acid:

$$\frac{dM_9}{dt} = \left(\frac{\dot{M}_{acid}}{m_{acid}}\right) + \left(\frac{Xm_{total} \times M_4}{P_{total}}\right) - \left(\frac{Xm_{2total} \times M_9}{P_{total}}\right) \times \left(\frac{R_{gás} \times T_4}{V_{gás}}\right) + \left(\frac{M_9 \times \frac{dT_4}{dt}}{T_4}\right) \quad (59)$$

$$M_{1gás1} = \frac{M_8 \times V_{gás} \times m_{dode}}{R_{gás} \times T_4} \quad (60)$$

$$M_{2gás2} = \frac{M_9 \times V_{gás} \times m_{acid}}{R_{gás} \times T_4} \quad (61)$$

$$C_4 = (M_{1gás1} \times C_{p4dode}) + (M_{2gás2} \times C_{p4acid}) \quad (62)$$

$$P_{3,2} = P_{total} - M_8 - M_9 \quad (63)$$

Balance of energy in the gas above the plate:

$$\frac{dT_4}{dt} = \frac{(M_{2dode} \times C_{p4dode} \times T_4) + (M_{2acid} \times C_{p4acid} \times T_4) + (Xm_{total} \times m_{dode} \times M_3) + \frac{P_{3,2} \times C_{p2dode} \times T_3}{P_{total}}}{C_4} + \quad (64a)$$

$$+ \frac{Xm_{total} \times m_{acid} \times M_4 \times C_{p2acid} \times T_3 - (Xm_{2total} \times m_{dode} \times M_8) + \frac{P_{3,2} \times C_{p4dode} \times T_4}{P_{total}} - \frac{(Xm_{2total} \times m_{acid} \times M_9 \times C_{p2acid} \times T_4)}{P_{total}}}{C_4} \quad (64b)$$

Runge-Kutta (Vargas & Araki, 2017) method was programmed in a FORTRAN<sup>®</sup> language program for the differential equation solutions. The initial model conditions, parameters and values for simulation are detailed in Tab. 1.

Table 1. Initial values used in the mathematical model.

Symbols	Description	Numerical Values	Units
$\rho$	Density	800.	kg.m <sup>-3</sup>
Adode	Antoine equation (dodecane)	4.10549	Dimensionless
Bdode	Antoine equation (dodecane)	1625.928	K
Cdode	Antoine equation (dodecane)	-92.839	K
Aac	Antoine equation (palmitic acid)	5.3573	Dimensionless
Bac	Antoine equation (palmitic acid)	3061.422	K
Cac	Antoine equation (palmitic acid)	-55.0770	K
Acp dode	Joback method for specific heat (Cp)	-8.02	kJ.mol <sup>-1</sup>
Bcp dode	Joback method for specific heat (Cp)	1.14384	kJ.mol <sup>-1</sup> .K <sup>-1</sup>
Ccp dode	Joback method for specific heat (Cp)	-0.000629	kJ.mol <sup>-1</sup> .K <sup>-2</sup>
Dcp dode	Joback method for specific heat (Cp)	0.0000001316	kJ.mol <sup>-1</sup> .K <sup>-3</sup>
Acp acid	Joback method for specific heat (Cp)	-7.056	kJ.mol <sup>-1</sup>
Bcp acid	Joback method for specific heat (Cp)	1.57462	kJ.mol <sup>-1</sup> .K <sup>-1</sup>
Ccp acid	Joback method for specific heat (Cp)	-0.0009192	kJ.mol <sup>-1</sup> .K <sup>-2</sup>
Dcp acid	Joback method for specific heat (Cp)	0.0000002072	kJ.mol <sup>-1</sup> .K <sup>-3</sup>
Kla dode	Mass transfer coefficient	0.0005	s <sup>-1</sup>
Kla acid	Mass transfer coefficient	0.0005	s <sup>-1</sup>
Vres	Reservoir volume	1.	m <sup>3</sup>
R	Universal gas constant	8.314x10 <sup>-2</sup>	kJ.mol <sup>-1</sup> .K <sup>-1</sup>
Mdode	Dodecane molar mass	170.	g.L <sup>-1</sup>
Macid	Palmitic acid molar mass	256.	g.L <sup>-1</sup>
Qres	Power dissipated by resistance	200000.	Watt
Ptotal	Total pressure	1.	atm
Vgas	Gas volume	20.1	m <sup>3</sup>
Vplate	Plate volume	0.05	m <sup>3</sup>

Kladodedish	Mass transfer coefficient	0.00005	s <sup>-1</sup>
Klaaciddish	Mass transfer coefficient	0.00005	s <sup>-1</sup>

Source: The Authors (2017).

#### 4. RESULTS AND DISCUSSION

According to Fig. 2, it can be seen that the mass decreases over time, demonstrating that a distillation is occurring. The initial mass of the flask is heated and the compound evaporates, fractionating the sample according to its boiling point of each compound.

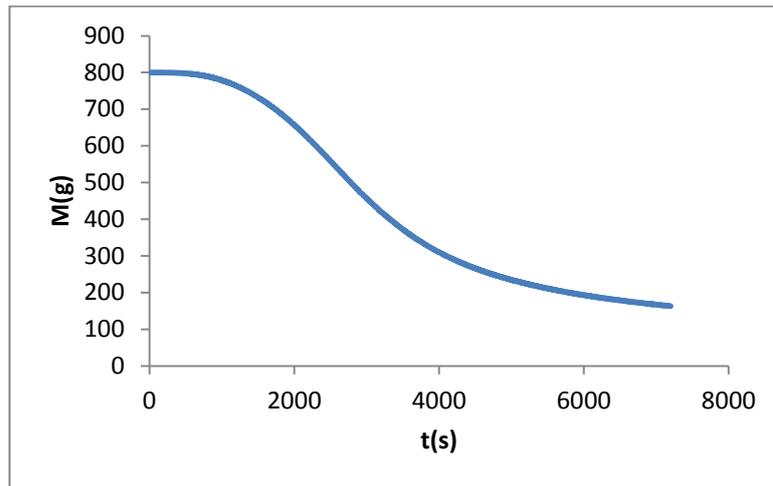


Figure 2. Decrease of mass according to time. Source: The Authors (2017).

The Fig.3 shows the behavior of the temperature in relation to time in the plate of the distillation column. After a certain amount of time the temperature drops to show that the plate of the column is full, at that moment the plate begins to overflow with a cooling of the compound.

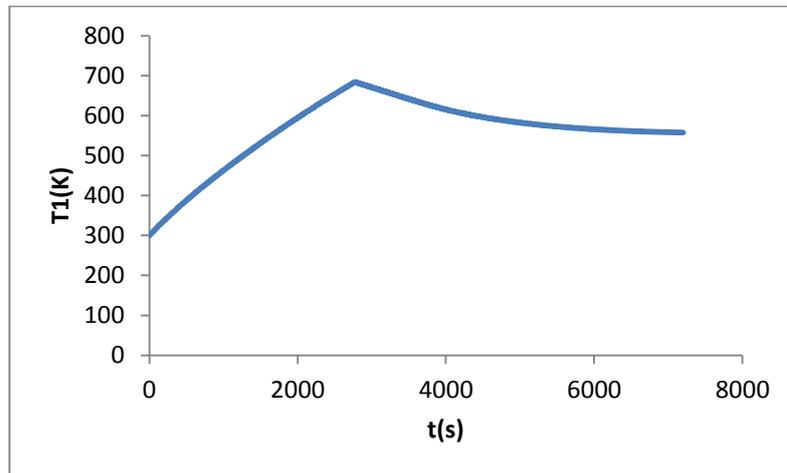


Figure 3. Behavior in the column plate. Source: The Authors (2017).

The Fig. 4 shows the sample distillation behavior, which is composed of dodecane and palmitic acid. Over time the compounds are separated, dodecane having a lower boiling point comes out first, which is the most interesting compound, about 60% of the sample is concentrated in dodecane. Palmitic acid is heavier than dodecane and is a compound that does not concern the fractional distillation of microalgae hydrocarbons.

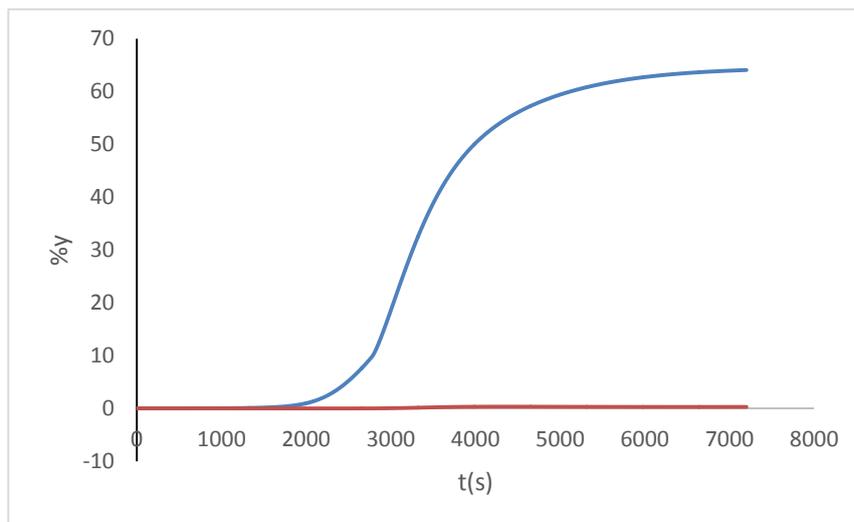


Figure 4. Fractionation of compounds. Source: The Authors (2017).

The Fig. 5 shows the behavior of the palmitic acid in the fractional distillation, it is not noticed change in its volume over the time, because the temperature used in this model was not high enough for the palmitic acid to leave in greater quantity. It is important to ensure a temperature where this compound is not fractionated, as it is not of interest.

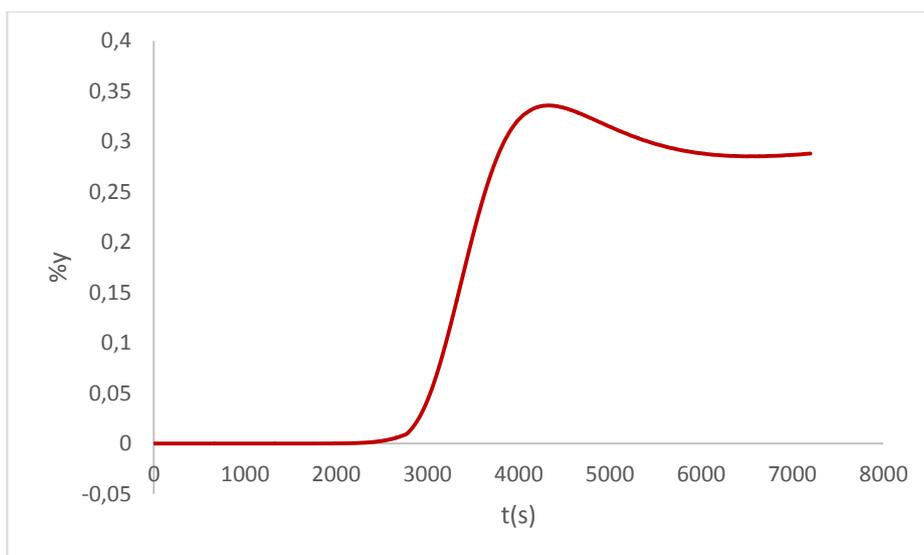


Figure 5. Behavior of palmitic acid. Source: The Authors (2017).

After fractional distillation it was necessary to characterize the sample; this characterization was done by gas chromatography with mass spectrometry. The chromatogram as shown in Fig. 6 was performed at the Chemistry Laboratory of the Chemistry Department of UFPR.

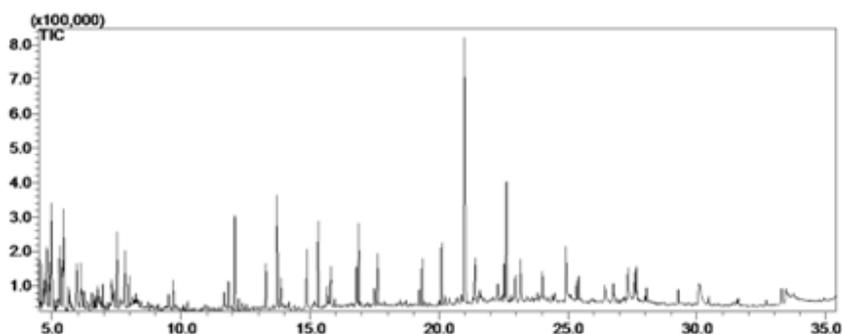


Figure 6. Chromatogram. Source: The Authors (2017).

The analysis shows the presence of hydrocarbons of 12 to 19 carbons, which can be used as fuel. Tab. 2. shows the compounds present in the distilled sample.

Table 2. Percentage of microalgas extract components by area standardization.

Retention time (minutes)	Structure Possibility	Area	Percentage (%)
11,66	Tetradec-3-ene	92104	0,8857
11,83	Dodecan	153847	1,4795
12,08	2,5-dimethyl-undecane	539408	5,1874
12,23	Tridec-1-ene	67295	0,6471
13,28	2,6,10-trimethyl-dodecane	263652	2,5355
13,72	1,1,2-trimethyl-cycloundecane	641312	6,1674
13,87	Tridecane	166130	1,5976
14,87	3,7,11-trimethyl-dodecan-1-ol	296408	2,8505
15,31	2,6,10,14-tetramethylpentadecane	475474	4,5725
15,65	Hexadec-3-ene	95544	0,9188
15,79	Tetradecane	229447	2,2065
16,79	Nonadecane	232531	2,2362
16,89	Hexadecane	480901	4,6247
17,49	Pentadec-1-ene	110510	1,0627
17,62	Pentadecane	285508	2,7457
19,23	Heptadec-3-ene	99364	0,9555
19,34	Hexadecane	274210	2,6370
20,09	2,6,10-trimethylpentadecane	456128	4,3865
20,98	Heptadecane	2128254	20,4672
21,01	2,6,10,14-tetramethylpentadecane		
22,52	Octadecane	211122	2,0303
22,61	2,6,10,14-tetramethylhexadecane	945504	9,0928
22,96	3,7,11,15-tetramethylhexadec-2-ene	288321	2,7727
23,15	3,7,11,15-tetramethylhexadec-2-ene	300909	2,8938
24	Nonadecane	145039	1,3948
24,92	Hexadecanoic acid	367931	3,5383
25,34	Hexadecanoate of ethyl	109523	1,0532
25,41	Eicosano	128541	1,2361
27,32	Hexadec-9-enoic acid	279387	2,6868
27,57	Ethyl linoleate	154459	1,4854
27,65	Ethyl oleate	205284	1,9742
28,04	Docosano	86973	0,8364
29,28	Tricosano	87318	0,8397

Source: The Authors (2017).

Compounds with retention time of 16.79 minutes (Nonadecane) and 16.89 (Hexadecane) minutes, although having high similarity with the proposed compounds do not follow the elution logic of the sample, it is possibly another structure. Looking again at the spreadsheet we can see that there is again the suggestion of Hexadecane (19.34 min) and Nonadecane (24 min), having a greater probability of success due to the elution sequence of the hydrocarbon chains. It is observed that in the retention times of 22.96 and 23.15 minutes the possibility of having the same structure is high, since the spectrum is very similar. Simulations of the model will be presented in the complete paper.

## 5. CONCLUSION

The mathematical model met the objectives of this work, which was to show the fractional distillation of a sample with different compounds (dodecane and palmitic acid) and to analyze the behavior of each of them within the column in relation to the time and temperature at which the compound was left due to their respective boiling point. More compounds can be added to the model so that you can analyze the behavior more accurately. The model must be validated experimentally.

## 6. ACKNOWLEDGEMENTS

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