



HAL
open science

Experimental study on the effects of feedstock on the properties of biodiesel using multiple linear regressions

Aulia Qisthi Mairizal, Sary Awad, Cindy Rianti Priadi, Djoko Hartono, Setyo Moersidik, Mohand Tazerout, Yves Andres

► To cite this version:

Aulia Qisthi Mairizal, Sary Awad, Cindy Rianti Priadi, Djoko Hartono, Setyo Moersidik, et al.. Experimental study on the effects of feedstock on the properties of biodiesel using multiple linear regressions. Renewable Energy, Elsevier, 2020, 145, pp.375-381. 10.1016/j.renene.2019.06.067. hal-02269913

HAL Id: hal-02269913

<https://hal-imt-atlantique.archives-ouvertes.fr/hal-02269913>

Submitted on 25 Oct 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial| 4.0 International License

EXPERIMENTAL STUDY ON THE EFFECTS OF FEEDSTOCK ON THE PROPERTIES OF BIODIESEL USING MULTIPLE LINEAR REGRESSIONS

Aulia Qisthi Mairizal^{1,2}, Sary Awad^{1*}, Cindy Rianti Priadi^{2,3}, Djoko M. Hartono², Setyo S. Moersidik², Mohand Tazerout¹, Yves Andres¹

¹GEPEA, UMR-CNRS Department of Energy System and Environment (DSEE) IMT Atlantique, Nantes 44300, France

²Environmental Engineering Study Program, Department of Civil Engineering, Faculty of Engineering, Universitas Indonesia, Depok 16424, Indonesia

³Tropical Renewable Energy Center (TREC), Faculty of Engineering, Universitas Indonesia, Depok 16424, Indonesia

* Corresponding author: sary.awad@gmail.com

Abstract. Biodiesel is a very promising alternative fuel that has its place in the future energy mix. The dependence of fuel properties on fatty acids profile will influence the choice of feedstock or appropriate treatment that it should undergo in order to respect biodiesel standards. The objective of this study is to find models that predict biodiesel's viscosity, density, flash point, higher heating value, and oxidative stability based on saponification value, Iodine value and the polyunsaturated fatty acids content of feedstock. Biodiesel samples were produced from seventeen different blends of oils. Multiple linear regressions were used to obtain models. High accuracy prediction was obtained for density and higher heating value with prediction errors < 5%, a very good accuracy was obtained for viscosity with error < 10% and flash point and oxidative stability were predicted with a fair accuracies (error < 15%) which indicates a good correlation level with IV, SV and Polyinsaturations but it also reveals that other parameters could also interfere and should be taken in consideration to reach acceptable accuracy.

Keywords: Iodine Value; Saponification Value; polyunsaturated fatty acid, Multiple linear Regressions

1. Introduction

29 The increasing energy demand, combined with declining global environmental conditions has
30 led to a shift towards utilization of more sustainable sources of energy. This shift signifies that
31 energy of the future may be highly dependent on sources that are environment-friendly but
32 highly variable. Biodiesel is a renewable alternative fuel that is biodegradable and has similar
33 properties with conventional diesel fuel [1]. Many researchers have identified biodiesel as a
34 good alternative fuel with high potential since it has a good balance in terms of environmental,
35 economic development and technical availability. As the demand of alternative fuels keeps
36 rising, biodiesel holds a lot of promise for future of transportation [2].

37 Biodiesel is defined as the mono alkyl esters of fatty acids derived from vegetable oils or animal
38 fats. Biodiesel could be produced from a large variety of feedstock. The most common
39 feedstock used for biodiesel production comes from vegetable oils and animal fats that are
40 mostly composed from triacylglycerol (TAG). In general, the TAG of vegetable oils and animal
41 fats are composed of different combinations of fatty acids (FA) having a wide variety in terms
42 of physical and chemical properties [3]. The main sources of properties variations are attributed
43 to the degree of unsaturation (it might be expressed in terms of iodine value) and the carbon
44 chain length (it might be expressed in terms of saponification value) [4]

45 Making mathematical correlations between feedstock composition from a side and quality
46 parameters on the other side, with a reasonable accuracy, could have several benefits on
47 scientific plan. For example, it could be a good estimation of the ability of a feedstock to
48 produce a good quality of biodiesel before undergoing time and money consuming
49 characterization by only using simple tests. At the same time, it could help to determine
50 appropriate treatment strategies of feedstock to improve biodiesel properties. These treatments
51 could be as simple as mixing two types of oil or advanced as hydrogenation [5] where a good
52 balance between saturated/unsaturated fatty acids is needed in order to meet norms. At the same

53 time, correlations could reduce the number of variables that could be used to define a biodiesel
54 in combustion engines simulations.

55 Several studies have demonstrated the influence of fatty acids methyl esters (FAME) profile on
56 biodiesel properties. Allen et al. (1999) [6] proposed a quadratic model to predict viscosity from
57 fatty acid composition. Ramírez-Verduzco et al. (2012) [7] developed a correlation to estimate
58 cetane number, viscosity, density and higher heating value as a function of iodine value (IV)
59 and molecular weight. Pinzi et al. (2011) [8] proposed a mathematical model for low calorific
60 value, kinematic viscosity, flash point, cetane number and cold filter plugging point with the
61 independent factors of IV and carbon chain length. Ramos et al. (2009) [9] predicted cold filter
62 plugging point (CFPP) using chain length and saturation factor, while Yuan et al. (2017) [10]
63 presented the relation between CFPP and FAME. Sarin et al. (2010) [11] performed a
64 mathematical model for oxidation stability as a function of unsaturation degree and palmitic
65 acid content. Lapuerta, et al. (2009) [12] reported the estimation of cetane number of biodiesel
66 as a function of iodine value and number of carbon atoms.

67 All works found in literature dealt with unsaturation level using IV, but no one has investigated
68 the effect of mono and polyunsaturated fatty acids balance (PU/MU) on the characteristics of
69 biodiesel. Thus, the present work aims to introduce new correlations that include, besides SV
70 and IV, the PU/MU ratio as an independent parameter in order to predict viscosity, density,
71 flash point (FP), higher heating value (HHV), and oxidative stability (OS) with fair precisions.
72 To do so, wide ranges of IV, SV and PU/MU profiles were investigated. The IV ranged from
73 (0 - 148 gI₂/100g_{oil}), SV ranged from (188-265 gKOH/g_{oil}) and PU/MU ranged between (0 – 3.87).
74 Those ranges were reached by using blends of seven different types of feedstock. A multiple
75 regression analysis was carried out in order to determine correlations and analysis of variance
76 (ANOVA) was derived in order to study the significance of correlations.

77 All the oils were characterized by gas chromatography-mass spectrometry (GC-MS) to find the
78 fatty acid methyl ester (FAME) profile and then tested according to European biodiesel standard
79 EN 14214.

80 **2. Materials and Methods**

81 *2.1. Raw Materials*

82 Seven different types of oil were used during this study. Sunflower oil, peanut oil, hydrogenated
83 coconut oil, hydrogenated copra oil, beef tallow, rapeseed oil and walnut oil. These types of oil
84 were purchased from local stores in Nantes, France. Methanol and potassium hydroxide were
85 purchased from Sigma Aldrich Company.

86 *2.2. Transesterification Process*

87 The biodiesel production was carried out with transesterification process by using an alkali
88 catalyst KOH and methanol in a 1 L flat bottom flask. The process was accomplished under the
89 following conditions: 6:1 methanol/oil molar ratio, 400 rpm rotational speed, 50 °C reaction
90 temperature, 1 wt.% catalyst dosage based on oil weight, and a reaction time of 2 hours.

91 At the end of transesterification process, the samples were left overnight to settle the phase
92 separation between glycerol and crude methyl ester. Then, the lower layer containing the
93 glycerol and other impurities was removed. After that, the crude methyl ester was washed a few
94 times with warm distilled water at 50°C until the pH of last washing water became neutral. The
95 residual water and methanol in the mixture were separated from biodiesel product with rotary
96 evaporation under vacuum at 40°C for 1 hour. Finally, the yield of biodiesel was measured at
97 this step and after that, samples were characterized.

98 *2.3. Blend Preparation*

99 Feedstock (7 different types of oil) were chosen carefully in order to present a wide range of
100 SV and IV and different balances between monounsaturated and polyunsaturated fatty acids.
101 Then blends were prepared from binary and ternary mixes of produced biodiesel.

102 *2.4. Analyses and Instruments*

103 To determine the FAME profile from various feedstock, 25 mg biodiesel samples were injected
104 into a gas chromatograph Perkin Elmer Clarus 680 equipped with a flame ionization detector
105 to obtain the chromatogram and peak integration report. Iodine value was calculated using the
106 following expression developed by Knothe (2002) [13]:

$$107 \quad IV = 100 \times \sum \frac{(\% FA) \times 253.81 \times db}{MW} \quad (\text{eq 1})$$

108 Where, *IV* is the iodine value of the oil, *db* is the number of double bonds per FA molecule,
109 *MW* is the molecular weight of each fatty acid and *% FA* is percentage of each fatty acid in oil.
110 The calculation was also conducted to determine the saponification value of each biodiesel
111 produced, where *SV* is the saponification value of the oil, with the equation 2 [13]:

$$112 \quad SV = 100 \times \sum \frac{(\%FA) \times 56.106}{MW} \quad (\text{eq 2})$$

113 *Biodiesel Characterization*

114 HHV analysis was carried out following the ASTM D3180 Standard, using Parr 6200
115 Calorimeter The results were expressed in MJ/kg with a relative error of 0.25%. OS was
116 determined with PetroOxy device following the ASTM D7525 standard.

117 Densities of different samples were measured at 15°C using a pycnometer M50T (850 – 900 g/l)
118 with a precision of 1 g/l. While an AND vibro viscometer was used to measure the dynamic
119 viscosity at 40°C, then kinematic viscosity was obtained by dividing it by the density. The
120 relative error of kinematic viscosity was estimated to 3%.

121 Flash point was measured using a PENSKY MARTENS NPM440 device with a precision of
122 1°C.

123 2.5. *Statistical analysis*

124 A multiple regression analysis was conducted in order to describe the relationship between each
125 of the independent variables (SV, IV and PU/MU) and the dependent variables of the samples
126 (characteristics matrix). Moreover, ANOVA was performed for each regression in order to
127 determine its significance and the significance of each parameter. Finally, correlations were
128 tested by plotting predicted data versus experimental results. Models were then compared to
129 experimental data found in the literature.

130

131 **3. Results and Discussion**

132 3.1. Fatty Acid Methyl Ester Profile (FAME)

133

134 The fatty acid profile of different methyl esters is summarized in Table 1. The present work
135 shows that biodiesel derived from sunflower oil was very rich in unsaturated acids. Based on
136 experimental results, it contains high amounts of linoleic acid (C18:2) and oleic acid (C18:1).
137 Their fractions are 60 % and 29% respectively. Rapeseed oil has also high amount of
138 monounsaturated acids, which achieved 62 %, consisting mainly of oleic acid (C18:1). Both
139 sunflower and rapeseed oils are the most common used feedstock in biodiesel production in
140 EU.

141 The biodiesel derived from walnut oil has the highest percentage of polyunsaturated fatty acids
142 with 56 % linoleic acid (C18:2) and 11.3% of linolenic acid (C18:3). Beef tallow is composed
143 of a high variety of fatty acids with high concentrations of palmitic (33.72%) and stearic
144 (18.9%) acids which were the highest among the other investigated biodiesel samples. The same
145 result was also reported by Giakoumis (2013) [14].

146 Peanut oil is also one of the interesting feedstock to investigate since its profile covers very long
147 carbon chains reaching C24. Both methyl esters from hydrogenated coconut oil and
148 hydrogenated copra oil represent a short carbon chains compared to other feedstock (from C6
149 to C16).

150 3.2. Correlations between biodiesel properties and independent parameters

151 The study was carried out using a set of 17 experimental points (7 oils and 10 blends), covering
152 wide ranges of SV, IV and PU/MU ratio. The characteristics of experimental points are listed
153 in table 2.

154 These data were used to obtain wanted correlations using multiple linear regressions and results
155 are listed in table 3, while ANOVA is listed in table 4.

156 3.2.1. Density

157 In general, density of biodiesel (860-900 kg/m³) is slightly higher than that of petroleum diesel
158 (820-845 kg/m³). Density has an important role, especially during the fuel injection, since a
159 higher fuel density will lead to a higher fuel mass injection in the engine. Therefore, the energy
160 content within the combustion chamber and the engine performance are highly influenced by
161 fuel density [1].

162 In the US, there is no specification for biodiesel density, but in European biodiesel standard,
163 EN 14214, it is mentioned that the acceptable range of density lies between 860-900 kg/m³
164 while in Indonesian National Standard (SNI 7182:2015) the range of density lies between 850-
165 890 kg/m³. The main reason behind this limitation is to avoid the significant amount of
166 polyunsaturated fatty acids in the fuel [15].

167 Based on the experimental results presented in table 2, the densities from all investigated
168 biodiesel samples, range from 862.9 kg/m³ to 877.6 kg/m³ with an overall average value of
169 870.9 kg/m³. The results show that all examined methyl esters meet both the EU and Indonesian
170 standards specifications. Highest density (877.6 kg/m³) was registered for biodiesel derived
171 from walnut oil. This amount resulted from its high content of unsaturated fatty acids (84.7%)

172 with a high balance of polyunsaturated fatty acids (PU/MU = 3.87). This result is in line with
173 the investigations led by Ramírez-Verduzco et al. (2012) [7] that biodiesel rich in unsaturated
174 compound such as linoleic acid (C18:2) and oleic acid (C18:1) will have a higher density.

175 As it is listed in table 3 a model's fit between density as a function of IV, SV and PU/MU ratio
176 was developed and showed high coefficient of determination ($R^2= 95.4\%$) using a quadratic
177 model. The relative error between measured and predicted data was lower than 0.3%, as it is
178 shown in Figure 1. Lowest p -values were found for IV, IV^2 and $IV*SV$ which reflects the strong
179 correlation between the degree of unsaturation with the value of density, which seems to be
180 more significant than the carbon chain length and the balance between mono- and poly-
181 unsaturated fatty acids. Nevertheless, including PU/MU into the correlations have decreased
182 standard error from 6 kg/m^3 to 1.52 kg/m^3 and increased the significance of the correlation.
183 Giakoumis (2013) [14] reported that density would increase with increasing unsaturation
184 degree. This is in line with study led by Ramírez-Verduzco et al. (2012) [7] that density is
185 directly proportional with degree of unsaturation with the additional increase of density 0.00118
186 g/cm^3 for each additional double bond.

187 In figure 1, the model was also compared to the data from literature, it can be noticed that the
188 maximum relative deviation between experimental data and predicted values was around 2.2%
189 while the majority of points lie between the $\pm 2\%$ relative error limits. It could also be noticed
190 that all data reported by Giakoumis et al. [14] and Yuan et al. [10] lie under the $Y=X$ line, which
191 means that the model tends to underestimate the density values. Taking into account the low
192 error margin, this could be related to the methods used for density measurements and the
193 apparatus related errors.

194 3.2.2. Higher Heating Value

195 Heating value is an important fuel property since it defines the amount of energy that will be
196 released during FAME combustion in the engine. The heating value is also known as the heat

197 of combustion and could be divided into two types as lower heating value (LHV) and higher
198 heating value (HHV). Both are the measurement units to indicate the heat of combustion when
199 the fuel is burned completely. It is expressed as a unit of energy released per quantity of the fuel
200 (MJ/kg). LHV is deduced from HHV by subtracting the heat of vaporization of water formed
201 during combustion. The HHV could be explained as a function of hydrogen content, carbon
202 content and oxygen content with equation performed by Demirbas (1998) [16]. There is no
203 specific limit of higher heating value mentioned in European biodiesel standards, US ASTM D
204 6751-08 and Indonesian National Standard (SNI 7182:2015). However, due to its significant
205 oxygen content (10-12% w/w) [14], it is generally expected that the energy content of biodiesel
206 will be lower than diesel fuel [1, 7, 17]. As a result, higher fuel injection rates are required,
207 when engines are fuelled with biodiesel, in order to deliver power outputs similar to those
208 obtained with diesel fuel [8, 14].

209 The results of this study show, as illustrated in Table 2, that saturated biodiesel, derived from
210 coconut oil, present a HHV of 38.43 MJ/kg and the most unsaturated methyl esters, derived
211 from walnut oil, recorded 39.6 MJ/kg. The highest SV of hydrogenated oils leads to a higher
212 concentration of oxygen, which reduces the HHV.

213 In the present work, the prediction of HHV was made as a function of SV and IV. The result of
214 the ANOVA analysis showed the best model that fits the experimental result was a linear model
215 with $R^2 = 76\%$. The values of experimental HHV and calculated ones are compared in Figure
216 2, and they present a maximum relative error lower than 1.3%.

217 Furthermore, the *p*-value of SV demonstrates a strong influence on HHV. The negative sign in
218 SV coefficient is in line with the earlier study by Demirbas (1998) [16] that demonstrated that
219 the decrease in SV will increase ratios of carbon and hydrogen to oxygen in fuels and increase
220 the specific heat of combustion. However, the positive sign of IV coefficient means that HHV
221 is increasing with unsaturation, which is in contradiction with the study of Ramírez-Verduzco

222 et al. (2012) [7] that showed HHV will decrease by 0.21 MJ/kg for each double bond in FAME
223 molecule. In fact, when IV increases, that means that the H/C ratio will decrease, leading to a
224 decrease in HHV. However, the energy content decrease due to the replacement of two
225 hydrogen atoms by a double bond in a molecule is very low. Thus, the final result could be
226 more influenced by other parameters such as the combustion efficiency in the calorimetric
227 bomb. So the lowest viscosity of unsaturated FAME could enhance mixing with air during
228 sample combustion and counterbalance the effect of energy content of hydrogen present in
229 saturated FAME.

230 Although the low R^2 recorded for the HHV, the comparison of the present model to other
231 researchers works showed that the maximum error was lower than 5%, with 86% of literature
232 data lying below 3% relative error, which reflects a good accuracy of the model.

233 3.2.3. Viscosity

234 Viscosity is a key biodiesel property, since it indicates the ability of a material to flow [17] and
235 has a strong relation with the behavior of fuel injection [18]. High viscosity of fuel will lead to
236 poor atomization and large droplet sizes of the fuel spray which leads to operational problems
237 [3]. European biodiesel standard EN 14214 has set an acceptable range of viscosity from 3.5 -
238 5.0 mm²/s, while US ASTM D 6751-08 accepts 1.9 - 6.0 mm²/s and Indonesian National
239 Standard (SNI 7182:2015) accepts 2.3 - 6.0 mm²/s.

240 As can be seen in Table 2, the viscosity values of all examined feedstock range from 2.63 to
241 4.75 mm²/s. The result is higher than the viscosity of common diesel fuel (1.3-2.4 mm²/s), due
242 to the large molecular mass and chemical structure of biodiesel [1].

243 Based on the experiments, methyl esters from hydrogenated coconut and copra oils have the
244 lowest viscosity (2.63 mm²/s). The outlier's value of viscosity from coconut oil was also found
245 by Giakoumis (2013) [14]. The reason behind this is that low values of kinematic viscosity are

246 obtained from biodiesel having short fatty acid chains [19]. In fact, the carbon chains found in
247 methyl esters from hydrogenated oils went until C16 (palmitic acid).

248 On the other hand, the highest value of viscosity was recorded for methyl esters derived from
249 peanut oil (4.75 mm²/s). This high viscosity comes from the high content of long carbon chains
250 FAME. The higher unsaturated-levels biodiesel will have lower values of viscosity. In the
251 present work, viscosities of biodiesel from different feedstock were measured at 40°C and
252 correlated to SV, IV and PU/MU. As shown in figure 3, the correlation found can predict
253 viscosity with a fair accuracy, where the observed relative error was lower than 6%.

254 Furthermore, the *p*-value of SV demonstrates that it has higher influence on viscosity than IV
255 and that the PU/MU ratio has negligible effect. The result is in line with the earlier studies that
256 demonstrated that viscosity increases with the increase of carbon chain length [16, 20]

257 The comparison of viscosity model to data from literature shows that the maximum deviation
258 is around 12% and around 90% of inspected data points lie between the ±10% relative error
259 lines. It is to be noted that most of the references cited viscosities values with measuring errors
260 exceeding 10% of measured values and which were excluded from comparisons. Taking in
261 counts the fair error of prediction, the present model can be reliable for biodiesel viscosity
262 prediction of FAME mixtures.

263 3.2.4. Flash Point

264 Flash point is defined as the minimum temperature of the fuel at which its vapors ignite in
265 presence of air and a heat source [21]. The minimum limits of flash point are 100°C, 120°C and
266 130°C for SNI 7182:2015, EN 14214 and ASTM D 6751-08 respectively. These requirements
267 were set in order to ensure that the produced biodiesel has been purified from the excess
268 methanol that could decrease the flash point [18]. This high flash point of biodiesel has
269 advantages in terms of storage and fire hazard perspectives.

270 The flash points of biodiesel samples examined in the present work ranged from 124°C to
271 181.5°C. All the results from table 2 show that the biodiesel produced from different feedstock
272 have met EU, US and Indonesian biodiesel standards.

273 Further study was conducted to propose mathematical prediction of flash point as a function of
274 IV, SV and PU/MU ratio. The best fit was found for a 3rd degree model as listed in Table 3 with
275 coefficient correlation $R^2 = 95.7\%$. The FP values predicted are compared to the measured ones
276 in Figure 4. The correlation found for this biodiesel property is fair since the maximum relative
277 error found is less than 7.5%. The results of this model were also compared to literature data
278 after filtration. In fact, all literature data that have standard deviations >10% were excluded
279 from the comparison. As it can be seen in figure 4, the maximum deviation between
280 experimental data and model prediction was lower than 15%, with 75% of compared data lying
281 between $\pm 14\%$ lines. This relatively high error could be referred to the fact that fatty acid
282 composition is not the only parameter that influences the flash point as claimed by Kumar
283 (2017) [22].

284 3.2.5. Oxidative Stability 285

286 Oxidative stability is one of the major issues for fuel storage. Generally, oxidation of fuels
287 occurs during storage under aerobic conditions. Earlier studies demonstrated that there is a link
288 between oxidation stability and the number of double bonds in FA structure [14, 18] that makes
289 biodiesel susceptible to degradation in the presence of oxygen [22]. Other factors also can
290 influence the oxidative stability as acid value, peroxide value, the presence of air, metals, heat,
291 light or pressure, and also polymer content [23]. However, the rate of oxidation could be slowed
292 down by adding antioxidants.

293 European norm EN 14214 has set the minimum value of oxidative stability for biodiesel to
294 8 hours by using the Rancimat method, while in US biodiesel standard US ASTM D 6751-08
295 the minimum level is 3 hours, and 8 hours for Indonesian National Standard SNI 7182:2015.

296 The PetroOxy method, used in this work, has the advantage of accelerating the oxidation
297 process which reduces drastically the time needed to achieve the analysis. Several studies have
298 reported correlations between PetroOxy number and Rancimat number, but there is still some
299 divergence between results. For example, by fitting the results of the present study to models
300 suggested by Botella et al. [24] and Neumann et al. [25], deviations reaching 240% were
301 observed. Thus, in the present work, it was opted to correlate PetroOxy number to oil
302 characteristics directly.

303 The oxidative stabilities of biodiesel samples examined in the present work covered a wide
304 range lying between 6.23 – 108 minutes. For example, the methyl esters from walnut oil,
305 sunflower oil and rapeseed oil which are rich in unsaturated fatty acids have the lowest oxidative
306 stabilities, around 6.2 min. From the experiments, it could be seen that there is a correlation
307 between unsaturated compounds content in methyl esters and oxidation. This is in line with
308 Shahabuddin et al. (2012) [26] that reported the oxidative stability will decrease with increasing
309 degrees of unsaturation.

310 On the contrary, saturated fatty acids such as those contained in coconut oil and copra oil are
311 way more stable, with the PetroOxy numbers reaching 108 minutes.

312 Further study was conducted in order to investigate the relation between PU/MU ratio with
313 oxidation stability. The study of oxidation stability as a function of IV and PU/MU ratio was
314 investigated. The SV was not included as an independent variable since the relation between
315 oxidative stability and SV was found very weak. A 3rd order model resulted in a correlation

316 coefficient R^2 of 94.1%. Figure 5 represents the predicted values of OS using the quadratic
317 model compared to measured ones and as it can be shown, the maximum error was around 13%.
318 Based on the p -value from ANOVA analysis, it was confirmed that the PU/MU (PU/MU and
319 (PU/MU)² terms) is the most influencing factor on the oxidative stability, which points out the
320 effects of polyunsaturations on the OS [27]. This observation comes in line with Kumar's work
321 [22] who reported that not only the presence of double bonds influences the rate of oxidation,
322 but also their position. Karavalakis et al. (2010) [28] also showed that polyunsaturated FAME
323 are more susceptible to oxidation degradation than monounsaturated, because of the number of
324 reactive bis-allylic, that is why polyunsaturation is more prone to autoxidation.

325 The value of R^2 and the relatively high deviation (13%) between experimental measurements
326 and predicted OS values could be explained by the fact that the oxidative stability is very
327 sensitive to other parameters such as moisture content, peroxide value, acid value, glycerides
328 content, etc. More parameters need to be investigated in order to get accurate prediction of this
329 fuel characteristic.

330 **4. Conclusion**

331 In the present work, multiple linear regressions were used to predict biodiesel viscosity, density,
332 flash point, higher heating value and oxidative stability as a function of Saponification value,
333 Iodine value and Polyunsaturated/Monounsaturated fatty acids balance (PU/MU). The
334 Investigations were carried out using seventeen blends derived from seven different oils. The
335 results showed good correlations for all parameters, excluding flash point and oxidative
336 stability. High accuracy was obtained for density and higher heating value, a very good one for
337 viscosity and a fair prediction was obtained for Flash Point. Adding PU/MU as an independent
338 parameter increased accuracy and correlation coefficients for prediction models.

339 **References**

- 340 [1] Mahmudul, H., Hagos, F., Mamat, R., Adam, A., Ishak, W., & Alenezi, R. Production,
 341 characterization and performance of biodiesel as an alternative fuel in diesel engines - A review.
 342 *Renewable and Sustainable Energy Reviews* 2017;72:497-509.
- 343 [2] Antolín, G., Tinaut, F., Briceno, Y., Castaño, V., Pérez, C., & Ramírez, A. Optimisation of
 344 biodiesel production by sunflower oil transesterification. *Bioresource Technology* 2002;83: 111-
 345 4.
- 346 [3] Knothe, G., Gerpen, J., & Krahl, J. *The Biodiesel Handbook*. Champaign, Illinois: AOCS
 347 Press;2005.
- 348 [4] Saraf, S., & Thomas, B. Influence of feedstock and process chemistry on biodiesel quality.
 349 *ICHEME* 2007;85(B5):360-4
- 350 [5] Fattah, I. R., Kalam, M., Masjuki, H., & Wakil, M. Biodiesel production, characterization, engine
 351 performance, and emission characteristics of Malaysian Alexandrian laurel oil. *RSC Adv.*,
 352 2014,4, 17787-17796.
- 353 [6] Allen, C., Watts, K., Ackman, R., & Pegg, M. Predicting the viscosity of biodiesel fuels from their
 354 fatty acid ester composition. *Fuel* 1999;78:1319-26.
- 355 [7] Ramírez-Verduzco, L., Rodríguez-Rodríguez, J., & Jaramillo-Jacob, A. Predicting cetane number,
 356 kinematic viscosity, density and higher heating value of biodiesel from its fatty acid methyl ester
 357 composition. *Fuel* 2012;91:102-11.
- 358 [8] Pinzi, S., Leiva, D., Arzamendi, G., Gandia, L., & Dorado, M. Multiple response optimization of
 359 vegetable oils fatty acid composition to improve biodiesel physical properties. *Bioresource*
 360 *Technology* 2011;102:7280-8.
- 361 [9] Ramos, M. J., Fernández, C. M., Casas, A., Rodríguez, L., & Pérez, Á. Influence of fatty acid
 362 composition of raw materials on biodiesel properties. *Bioresource Technology* 2009;100:261–8.
- 363 [10] Yuan, M.-H., Chen, Y.-H., Chen, J.-H., & Luo, Y.-M. Dependence of cold filter plugging point
 364 on saturated fatty acid profile of biodiesel blends derived from different feedstocks. *Fuel*
 365 2017;195:59-68.
- 366 [11] Sarin, A., Arora, R., Singh, N., Sarin, R., & Malhotra, R. Blends of biodiesels synthesized from
 367 non-edible and edible oils: Influence on the OS (oxidation stability). *Energy* 2010;35:3449-53.
- 368 [12] Lapuerta, M., Rodríguez Fernández, J. L., & Mora, E. F. Correlation for the estimation of the
 369 cetane number of biodiesel fuels and implications on the iodine number. *Energy Policy*
 370 2009;37:4337–44.
- 371 [13] Knothe, G. Structure indices in FA chemistry. How relevant is the iodine value? *JAOCS*
 372 2002;79(9):847-54.
- 373 [14] Giakoumis, E. G. A statistical investigation of biodiesel physical and chemical properties, and
 374 their correlation with the degree of unsaturation. *Renewable Energy* 2013;50:858-78.
- 375 [15] Knothe, G., & Razon, L. F. Biodiesel fuels. *Progress in Energy and Combustion Science*
 376 2017;58:36-59.
- 377 [16] Demirbas, A. Fuel properties and calculation of higher heating values of vegetable oils. *Fuel*
 378 1998;77:1117-20
- 379 [17] Atabani, A., Silitonga, A., Badruddin, I. A., Mahlia, T., Masjuki, H., & Mekhilef, S. A
 380 comprehensive review on biodiesel as an alternative energy resource and its characteristics.
 381 *Renewable and Sustainable Energy Reviews* 2012;16:2070–93
- 382 [18] Hoekman, S. K., Broch, A., Robbins, C., Cenicerros, E., & Natarajan, M. Review of biodiesel
 383 composition, properties and specifications. *Renewable and Sustainable Energy Reviews*
 384 2012;16:143-69.
- 385 [19] Gopinath, A., Puhan, S., & Nagarajan, G. Theoretical modeling of iodine value and saponification
 386 value of biodiesel fuels from their fatty acid composition. *Renewable Energy* 2009;34:1806-11.
- 387 [20] Demirbas, A. Relationship derived from physical properties of vegetable oil and biodiesel fuels.
 388 *Fuel* 2008;87:1743-8.
- 389 [21] Bajpai, D., & Tyagi, V. Biodiesel: Source, Production, Composition, Properties and Its Benefits.
 390 *Journal of Oleo Science* 2006;55(10):487-502.
- 391 [22] Kumar, N. Oxidative stability of biodiesel: causes, effects and prevention. *Fuel* 2017;190:328-50.
- 392 [23] Knothe, G. Some aspect of biodiesel oxidative stability. *Fuel Processing Technology*

- 393 2007;88:669-77.
- 394 [24] Botella, L., Bimbela, F., Martin, L., Arauzo, J., & Sanchez, J. L. Oxidation stability of biodiesel
395 fuels and blends using the Rancimat and PetroOXY methods. Effect of 4-allyl-2,6-
396 dimethoxyphenol and catechol as biodiesel additives on oxidation stability. *Frontiers in chemistry*
397 2014;2(43):1-9.
- 398 [25] A. Neumann, T. Jebens, V. Wiembicki, A Method for Determining Oxidation Stability of
399 Petrodiesel, Biodiesel, and Blended Fuels, *American Laboratory* 2008; 40(4): 22-26. Available
400 online at: [http://www.americanlaboratory.com/914-Application-Notes/34716-A-Method-for-](http://www.americanlaboratory.com/914-Application-Notes/34716-A-Method-for-Determining-Oxidation-Stability-of-Petrodiesel-Biodiesel-and-Blended-Fuels/)
401 [Determining-Oxidation-Stability-of-Petrodiesel-Biodiesel-and-Blended-Fuels/](http://www.americanlaboratory.com/914-Application-Notes/34716-A-Method-for-Determining-Oxidation-Stability-of-Petrodiesel-Biodiesel-and-Blended-Fuels/)
- 402 [26] Shahabuddin, M., Kalam, M., Masjuki, H., Bhuiya, M., & Mofijur, M. An experimental
403 investigation into biodiesel stability by means of oxidation and property determination. *Energy*
404 2012;44:616-22.
- 405 [27] Saluja, R. K., Kumar, V., & Sham, R. Stability of biodiesel - A review. *Renewable and Sustainable*
406 *Energy Reviews* 2016;62:866-81.
- 407 [28] Karavalakis, G., Stournas, S., & Karonis, D. Evaluation of the oxidation stability of
408 diesel/biodiesel blends. *Fuel* 2010;89:2483-9.
- 409
- 410

411

412 **Figures Captions**

413

414 Figure 1: Predicted versus measured density plot

415 Figure 2: Predicted versus measured HHV plot

416 Figure 3: Predicted versus measured Viscosity plot

417 Figure 4: Predicted versus measured Flash point plot

418 Figure 5: Predicted versus measured Oxidative stability plot

419

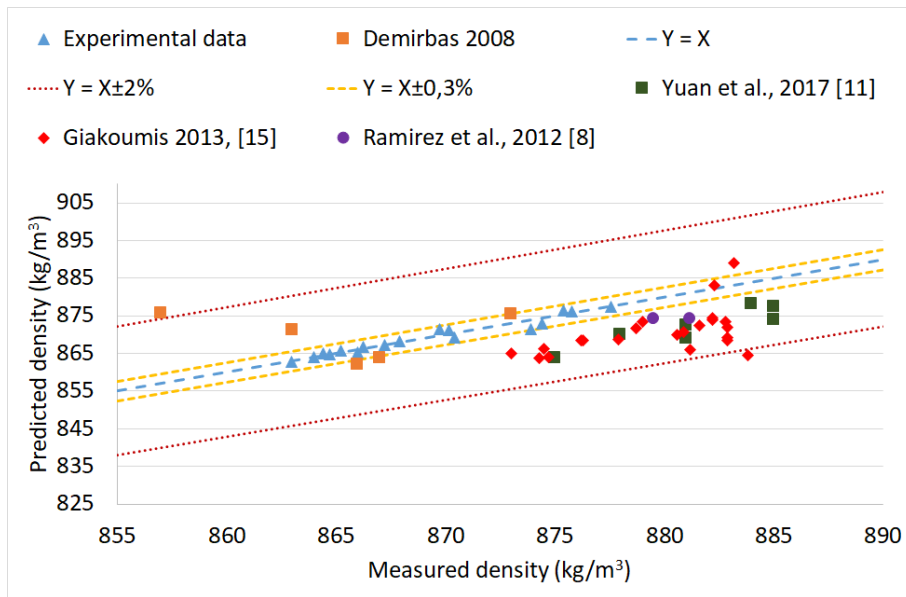


Figure 1: Predicted versus measured density plot

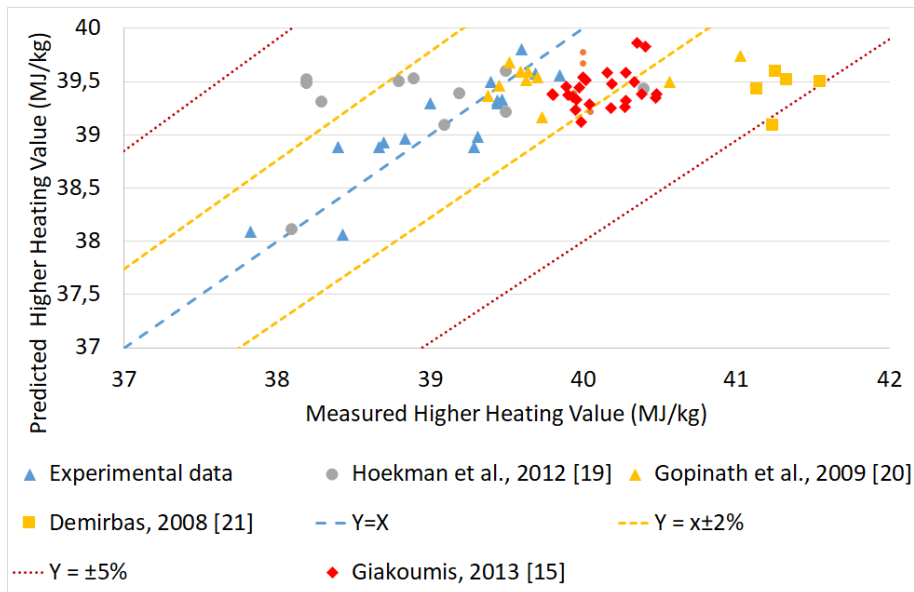


Figure 2: Predicted versus measured HHV plot

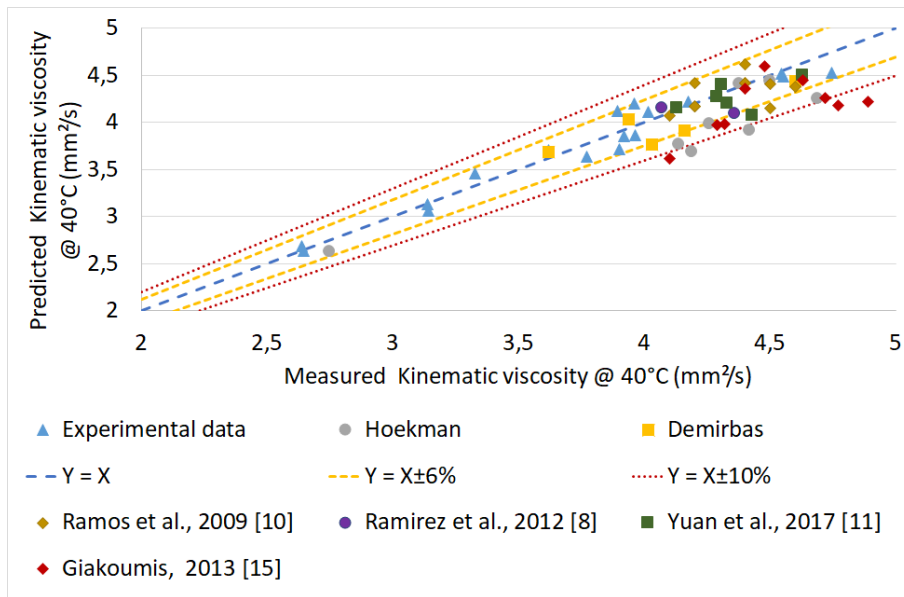


Figure 3: Predicted versus measured Viscosity plot

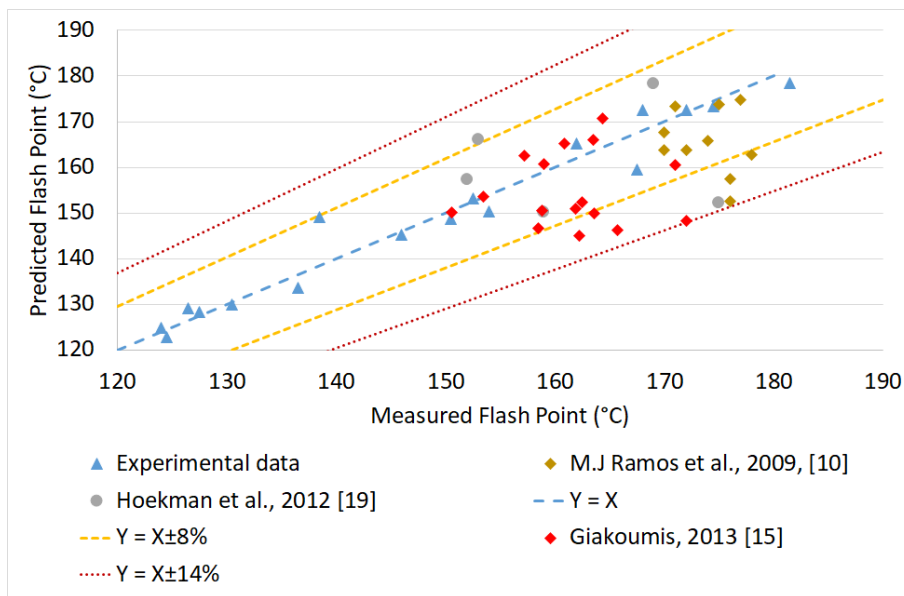


Figure 4: Predicted versus measured Flash point plot

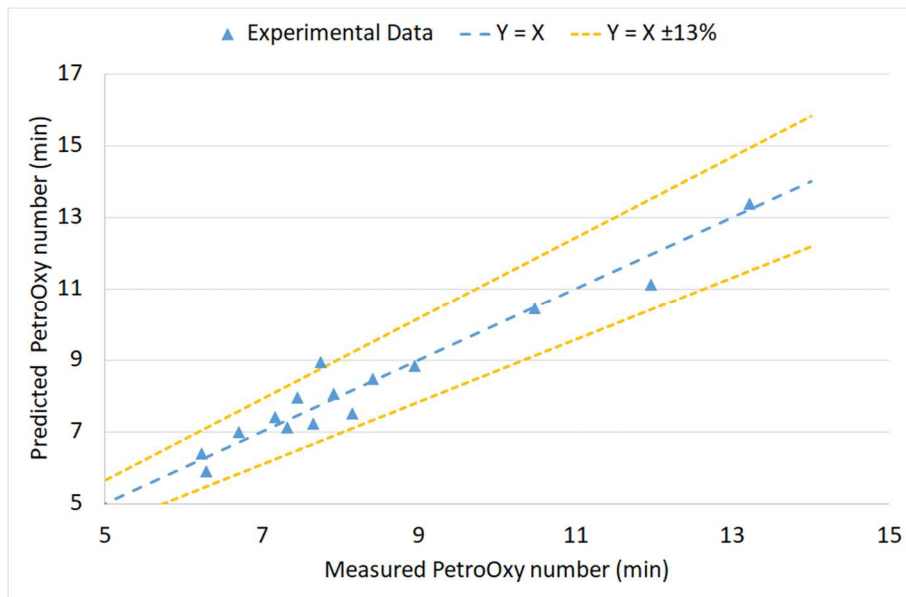


Figure 5: Predicted versus measured Oxidative stability

Table 1

Fatty Acid Composition (wt.%) of different types of oils

Carbon Chain	C6:0	C8:0	C10:0	C12:0	C14:0	C16:0	C18:3	C18:2	C18:1	C18:0	C20:1	C20:0	C22:0	C24:0
Molecular Weight	116	144	172	214	242	270	292	294	296	298	324	326	340	382
Formula	C ₇ H ₁₂ O	C ₉ H ₁₆ O ₂	C ₁₁ H ₂₀ O	C ₁₃ H ₂₆ O	C ₁₅ H ₃₀ O	C ₁₇ H ₃₄ O	C ₁₉ H ₃₂ O	C ₁₉ H ₃₄ O	C ₁₉ H ₃₆ O	C ₁₉ H ₃₈ O	C ₂₁ H ₄₂ O	C ₂₁ H ₄₂ O	C ₂₃ H ₄₄ O	C ₂₅ H ₅₀ O
	2		2	2	2	2	2	2	2	2	2	2	2	2
Sunflower Oil						6.5%		60.0%	29.0%	4.3%				
Peanut Oil						9.4%	18.0%		65.0%	3.6%			1.0%	1.6%
H.O. Coconut Oil	1.4%	8.5%	6.8%	50.3%	18.5%	8.2%				6.2%				
H.O. Copra Oil		3.8%	10.3%	54.5%	17.8%	7.5%				6.1%				
Rapeseed Oil						4.2%	8.4%	22.0%	60.0%	1.6%	2.1%			
Beef Tallow					4.3%	33.7%		0.8%	34.7%	18.9%	0.5%			
Walnut Oil						5.5%	11.3%	56.0%	13.8%	3.0%	3.6%	1.1%		

Table 2

Properties of biodiesel from various feedstock and mixtures

Parameter	IV	SV	PU/MU	Viscosity at 40°C	Density at 15°C	Flash Point	OS (Petrooxy)	Higher Heating Value
Unit	gI2/100g	gI2/100g	-	mm²/s	kg/m³	°C	min	MJ/kg
<i>Feedstock</i>								
Sunflower	134.981	200.29	2.07	3.96	875.78	174.5	6.3	39.69
Peanut	107.88	197.08	0.28	4.75	874.43	168	8.95	39.4
HO Coco	0.00	265.22	0	2.64	866	162	103.47	38.43
HO Copra	0.00	263.35	0	2.64	864.39	154	108.89	37.83
Beef tallow	33.19	192.96	0.024	4.55	862.94	172	13.22	39.2
Rapeseed	118.70	196.21	0.49	4.55	875.39	181.5	6.7	39.85
Walnut	147.95	188.40	3.87	3.97	877.58	152.5	6.23	39.6
Mixture 1	53.53	223.72	0.24	3.90	864	130.5	11.97	38.67
Mixture 2	36.49	217.83	0.39	3.62	864.69	127.5	10.48	39.29
Mixture 3	77.62	224.43	1.03	3.77	867.22	124	8.15	39.31
Mixture 4	62.83	226.54	2.49	3.14	870.43	126.5	7.92	38.4
Mixture 5	80.16	226.43	2.14	3.33	865.22	124.5	7.45	38.84
Mixture 6	73.85	226.93	3.81	3.14	866.22	146	7.32	38.7
Mixture 7	99.27	206.70	0.45	4.17	869.74	167.5	8.42	39.47
Mixture 8	63.75	197.40	0.59	3.89	867.88	150.5	7.75	39
Mixture 9	99.41	207.07	0.77	4.02	873.88	138.5	7.2	39.44
Mixture 10	84.38	204.21	1.23	3.92	870.14	136.5	7.65	39.44

Table 3
Summary of Models Developed to Predict Biodiesel Properties

Quality parameter	Units	Equation Model	R ²	Standard error
Density	kg/m³	$993 - 0.899.IV - 1.091.SV + 78.87.PU/MU + 32.5E-4.IV.SV - 0.189.IV.PU/MU - 0.29.SV.PU/MU + 28.4 E-4.IV^2 + 22.95E-4.SV^2 + 0.22.(PU/MU)^2$	95.4%	1.52 kg/m ³
Higher Heating Value	MJ/kg	$41.76 + 0.0045.IV - 0.0139.SV$	76%	0.29 MJ/kg
Viscosity (at 40°C)	mm²/s	$9.152 + 0.00572.IV - 0.0245.SV - 3.522.PU/MU - 1.37.E-5.IV.SV + 0.00533.IV.PU/MU + 0.013.SV.PU/MU$	95.5%	0.17 mm ² /s
Flash Point	°C	$-11749 - 4.364.IV + 173.76.SV - 8.07.PU/MU + 0.0276.IV.SV - 0.418.IV.PU/MU - 0.0149.SV.PU/MU - 0.0249 IV^2 - 0.833 SV^2 + 17.718 (PU/MU)^2 + 0.00016IV^3 + 0.00131SV^3 - 1.8135(PU/MU)^3$	95.7%	8°C
Oxidative Stability	mn	$15.157 - 0.0169.IV - 10.284.PU/MU + 0.0214.IV.PU/MU + 0.00084.IV^2 + 4.54.(PU/MU)^2 - 5.37E-6.IV^3 - 0.688 (PU/MU)^3$	94.1%	0.7 mn

Table 4
ANOVA of regressions

	SSR	SSE	SST	MSR	MSE	F	Significance F	Max Relative error	Average relative error
Density	337.9	16.18	354.095	37.545	2.312	16.24	6.7E-4	0.27%	0.084%
Higher Heating Value	3.558	1.111	4.67	1.78	0.085	20.880	8.85E-5	1.26%	0.61%
Viscosity at 40°C	5.8	0.27	6.09	0.027	0.97	35.4	3.57E-6	6.15%	2.7%
Flash Point	5701.8	256.2	5958.029	475.15	64.05	7.41	0.033	7.6%	1.85%
Oxidative Stability	54.15	3.35	57.5	0.48	7.74	16.16	7.9E-4	13.12%	4.4%