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Predicting the effect of pressure on biodiesel density at pressures of up to 200 MPa based on fatty acid alkyl ester profiles and density values at atmospheric pressure

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25

26 **1. Introduction**

27 Biodiesel is a renewable, biodegradable and non-toxic diesel engine fuel, designated B100
28 and simply defined by standard specifications (American Society for Testing and Materials
29 D6751 [1]). Derived from an increasing variety of raw resources such as vegetable oils,
30 animal fats and waste cooking oils and fats, biodiesel fuels are mixtures of mono-alkyl esters
31 of long chain fatty acids. The distribution of carbon chain lengths and the degree of
32 unsaturation of Fatty Acid Alkyl Esters strongly depends on the feedstock sources. The esters
33 can be produced from transesterification of fatty acids with any kind of alcohol so long as it
34 meets the requirements of the standard specifications. Consequently, the nature of the alkyl
35 ester (methyl ester, ethyl ester, etc.) and the fatty acid alkyl ester profile significantly differ
36 from one biodiesel to another. This difference in biodiesel composition has a direct impact on
37 the thermophysical properties of the fuel [2] and consequently influences engine efficiency
38 and the content of substances potentially harmful to the environment or human health in the
39 exhaust gases. Among fuel characteristics, volumetric property and its derivative with respect
40 to pressure appear to play a significant part in engine performance and fuel consumption.
41 Density acts on the conversion of volume flow-rate to mass flow-rate, and therefore affects
42 engine power [3]. It also has an impact on the size of the atomized fuel drops during the fuel
43 injection process. In addition, isothermal compressibility has a strong influence on fuel
44 injection timing [4,5]. Consequently, in order to optimize injection processes or design new
45 injection systems, it is essential to know biodiesel density and how it is affected by pressure
46 across the entire operating pressure range of the engine. Diesel engines, fuel injection systems
47 in particular, continue to evolve to improve overall engine performance. For better
48 combustion efficiency, fuels are now being exposed to very high pressures in common rail
49 direct injection systems, where the fuel is directly injected into the combustion chamber at

50 pressures of up to 200 MPa [6,7]. Although many studies [8-12] have been undertaken to
51 predict the density of biodiesels as a function of temperature at atmospheric pressure or even
52 at moderate pressures (up to 50 MPa) [13,15], few studies have been carried out at higher
53 pressures. Consequently, the aim of the present work is to establish a correlation to predict the
54 influence of pressure on biodiesel density up to 200 MPa across a wide temperature range
55 from 280 to 400 K based only on Fatty Acid Methyl Ester (FAME) or Fatty Acid Ethyl Ester
56 (FAEE) distribution profiles.

57

58 As biodiesel fuels are composed mainly of fatty acid alkyl esters ranging from methyl
59 caprate (MC10:0) to methyl lignocerate (MC24:0) [16], i.e. components belonging to the
60 same chemical family, excess volume is limited and biodiesel density and compressibility can
61 be predicted from the volumetric properties of its components, assuming an ideal solution.
62 Consequently, predicting the effect of pressure on biodiesel density amounts to correlating the
63 density of each FAME and FAEE as a function of pressure. With this aim in mind, a
64 Murnaghan equation [17] was considered here to represent the effect of pressure on the
65 volumetric properties of fatty acid alkyl esters up to 200 MPa; this equation involves only two
66 parameters, and appears as one of the most suitable for deriving compressibility from high-
67 pressure density data [18]. For the predominant saturated and unsaturated components of
68 biodiesels (i.e. laurate, myristate, palmitate, oleate and linoleate) these parameters can be
69 determined by fitting density data, abundant at pressures of up to 50 MPa, 100 MPa and even
70 up to 200 MPa. However, besides these predominant fatty acid alkyl esters, many biodiesels
71 contain non-negligible quantities of other saturated and unsaturated components that might
72 have an influence on density depending on the biomass feedstock used. The densities at
73 atmospheric pressure of some of these minority components have been measured and
74 compiled by Pratas et al. [19], but the effect of pressure on volumetric properties has received

75 little attention and no high pressure data are available. To make up for this absence of data,
76 the choice was made to set up a database from a group contribution method that would serve
77 to determine Murnaghan equation parameters for all the fatty acid alkyl ester components
78 commonly encountered in biodiesels, and therefore calculate the density of biodiesel as a
79 function of pressure based on its fatty acid alkyl ester profile. The predictive capacity of the
80 proposed method concerning the effect of pressure on biodiesel densities was tested against
81 the experimental high pressure density data available in the literature for biodiesels coming
82 from various feedstocks.

83

84

85 **2. Estimation of pure fatty acid alkyl esters densities**

86 *2.1 High pressure density data for pure fatty acid alkyl esters*

87 Predicting the influence of pressure on biodiesel density based on fatty acid alkyl ester
88 profiles requires knowing the liquid density of pure components over a wide pressure and
89 temperature range. With this aim in mind, high pressure density data available in literature as
90 well as their temperature and pressure ranges we listed in Table 1. It can be noted in this table
91 that the data span the temperature range from 270 to 470 K and the pressure range from 0.1 to
92 200 MPa but most of the data were obtained from an oscillating U-tube density meter at
93 pressures up to 70 MPa or up to 140 MPa. Some data were determined at higher pressures by
94 integrating speed of sound measurements. Finally, two datasets were obtained using a
95 flowmeter and a piezometer respectively. As can be observed in this table, there are very few
96 high pressure density data available despite the important role they play in designing high
97 pressure fuel injection systems. A total of 4350 experimental density values were found for
98 just 11 components, 7 of which belong to the FAME family and 4 to the FAEE family. As can
99 be seen, the data available mainly concern the saturated fatty acid alkyl esters ranging from

100 C10:0 to C18:0 and two unsaturated fatty alkyl esters, C18:1 and C18:2. Even then, the values
 101 are not equally distributed across the p, T domain. Most data concern the pressure range below
 102 100 MPa. Only few data are available up to 200 MPa. In addition to these components, most
 103 biodiesels contain non-negligible quantities of other saturated and unsaturated fatty acid alkyl
 104 esters. The densities at atmospheric pressure of some of these components were measured.
 105 Table 2 lists the components present in most biodiesels and the researchers who measured the
 106 densities of these components under atmospheric conditions. This table shows that the effect
 107 of temperature on the volumetric properties of long-chain saturated and polyunsaturated
 108 methyl esters have been extensively measured by Pratas et al. [19]. Consequently, density
 109 data obtained at atmospheric pressure across a wide temperature range are available for all
 110 components belonging to the fatty acid methyl ester family. Long-chain ethyl esters have
 111 received less attention and there is a total absence of data for some of them, such as ethyl
 112 gadoleate, behenate, erucate and lignocerate.

113

114 2.2 Representing the effect of pressure on volumetric properties

115 Many studies have been carried out with the aim of creating a model able to predict the
 116 volumetric behavior of liquid based on theoretical considerations, but none of them have
 117 produced a universal equation of state. Consequently, an empirical equation of state must be
 118 considered for describing the effect of pressure on liquid density. For that purpose, the
 119 following form of the Murnaghan equation [17] was used in this work:

$$120 \frac{\rho}{\rho_{atm}} = (1 + B\tilde{p})^{-C} \quad (1)$$

121 where B and C are empirical fitting parameters dependent of temperature and where \tilde{p}
 122 corresponds to the relative pressure: $p - p_{atm}$. The temperature dependences of B and C are
 123 described by the polynomial forms:

$$124 B = b_0 + b_1T + b_2T^2 \quad (2)$$

125 $C = c_0 + c_1 T$ (3)

126

127 This equation was chosen as it adequately represents density within the limits of the pressure
128 range investigated here. Moreover, it leads to a simple linear form of the tangent bulk
129 modulus:

130 $K = \rho \left(\frac{\partial p}{\partial \rho} \right)_T = -\frac{1}{BC} (1 + B\tilde{p})$ (4)

131 and consequently gives a reliable prediction of compressibility from density data:

132 $\kappa_T = -\frac{BC}{1+B\tilde{p}}$ (5)

133

134 The fitting parameters b_i and c_i must be obtained for each pure component by fitting the
135 Murnaghan equation to experimental data within the expected pressure and temperature
136 ranges of application of the proposed method: 0-200 MPa; 280-400 K. However, as
137 mentioned in the previous paragraph, the data are not equally distributed. The data of some
138 components such as myristate and palmitate (either methyl or ethyl) do not span the full p, T
139 range investigated due to the appearance of liquid-solid phase transitions at high pressures and
140 low temperatures. For other components, such as long-chain methyl esters, only data at
141 atmospheric pressure are available. Finally, for some components such as long-chain ethyl
142 esters there are simply no data at all. Consequently, the database needs to be completed by
143 predicting values for the missing data points. Fortunately, the simple molecular structures of
144 fatty acid methyl and ethyl esters have a limited number of functional groups, and the carbon
145 chain lengths of ester species included in the composition of biodiesel vary very little
146 whatever the feedstock source (C:10 to C:24). The group-contribution concept can therefore
147 be straightforwardly applied to estimate the missing density data. Indeed, results from
148 previous studies [40] concerning the densities of pure fatty acid alkyl esters under ambient
149 conditions show that group-contribution methods applied to volume, such as the GCVOL

150 model [68, 69], are able to estimate atmospheric pressure densities with deviations of the order
151 of 1.5% for FAEEs and of less than 1% in the case of FAME components.

152 The method consists in calculating the molar volume of a pure component under atmospheric
153 pressure by a simple summation of group volume increments:

$$154 \quad v_{atm}^*(T) = \sum_j N_j v_j(T) \quad (6)$$

155
156 where N_j is the number of j groups occurring in the pure component and v_j its volume
157 contribution. The method can be extended to predict the influence of pressure on density by
158 considering the same summation of volumes and using the Murnaghan equation to take into
159 account the effect of pressure on volume contributions:

$$160 \quad v^*(P, T) = \sum_j N_j A_j (1 + B_j \tilde{p})^{C_j} \quad (7)$$

161 where B_j and C_j are function of temperature according to eqs. 2,3 and A_j is a function of
162 temperature of the following form:

$$163 \quad A_j = a_{0,j} + a_{1,j}T + a_{2,j}T^2 \quad (8)$$

164 This equation involves 8 parameters for each functional group, i.e. $a_{0,j}$, $a_{1,j}$, $a_{2,j}$, $b_{0,j}$, $b_{1,j}$,
165 $b_{2,j}$, $c_{0,j}$, $c_{1,j}$ that were determined from fitting density data of pure components reported in
166 Tables 1 and 2.

167 A decomposition involving five functional groups was chosen here to represent FAME and
168 FAEE. Two were used to characterize the long alkyl chain (CH_3 and CH_2). The third group
169 ($\text{CH}=\text{CH}$) focused on the degree of unsaturation. And the last two groups were added to take
170 into account the ester contributions in FAME (CH_3COO) and FAEE ($\text{C}_2\text{H}_5\text{COO}$) respectively.

171 As can be seen in Table 1, the available data is distributed across the relevant p, T range for
172 each functional group. The a_i coefficients of each group were first determined from
173 experimental data taken from Table 2, except those of alkyl groups (CH_3 and CH_2) which
174 were taken from the GCVOL model [68] revised by Ihmels and Gmehling [69] as the present fit

175 give nearly the same a_i values for these groups. Parameters b_i and c_i were then determined by
 176 regression analysis of high pressure data reported in Table 1. The results of the regression are
 177 listed in Table 3.

178 The proposed group-contribution method helped fill in missing data and construct a
 179 comprehensive database with N_{exp} experimental data ranging from 0.1 to 200 MPa and from
 180 280 to 400 K. From this comprehensive database, coefficients a_i, b_i and c_i were estimated for
 181 each FAME and FAEE considered in Table 1. Parameters b_i and c_i were regressed from high
 182 pressure data by minimizing the following objective function:

$$183 \quad f_{HP}(b_i, c_i) = \sum_k^{N_{exp}} \left[(1 + B\tilde{p})^C - \left(\frac{\rho_{atm}}{\rho} \right)_{k,exp} \right]^2 \quad (9)$$

184

185 whereas a_i coefficients were independently fitted by minimizing the following objective
 186 function:

$$187 \quad f_{atm}(a_i) = \sum_k^{N_{exp,atm}} \left[A - \left(\frac{1}{\rho_{atm}} \right)_{k,exp} \right]^2 \quad (10)$$

188

189 The values resulting from the fit are given in Table 4. To study the capacity of eq. 1 to
 190 represent the effect of pressure on density with the fitted parameter values, the average
 191 deviations AD%, the absolute average deviations AAD% and the maximum deviations MD%
 192 between calculated and experimental values of the density ratio ρ/ρ_{atm} were evaluated for
 193 each experimental dataset. The deviations observed for each component are given in Table 5.
 194 These deviations were chosen as they are completely independent from data at atmospheric
 195 pressure, and therefore it corresponds to the use of the model for the purpose of extrapolating
 196 known data at atmospheric pressure to high pressure according to:

$$197 \quad \rho = \rho_{atm,exp} \left(\frac{\rho}{\rho_{atm}} \right)_{calc} = \rho_{atm,exp} (1 + B\tilde{p})^{-C} \quad (11)$$

198 leading to a relative uncertainty in terms of density corresponding to a quadratic summation
199 of relative uncertainties:

$$200 \quad U_r^2(\rho) = U_r^2(\rho_{atm,exp}) + U_r^2((\rho/\rho_{atm})_{calc}) \quad (12)$$

201 **3. Predicting the effect of pressure on the volumetric properties of biodiesels**

202

203 Most biodiesels are simple systems with few components that belong to the same family and
204 whose carbon chain length generally ranges from 10 to 20. Consequently, excess volumes in
205 biodiesels are very small and are often neglected when predicting density. In fact, the main
206 drawbacks when predicting densities from ester profiles are uncertainty in chemical analysis
207 and impurities. Indeed, end biodiesel products are obtained by conversion processes of
208 feedstock oils that leave several types of impurities. After transesterification of the fatty acids
209 with alcohol, the reaction product must be washed and purified to remove the glycerol and
210 unconverted fatty acids from the biodiesel phase. Given that the purification processes are not
211 100% efficient, some impurities remain in variable proportions in biodiesels depending on
212 their quality. Consequently, the molar volume of a biodiesel can be expressed as the sum of
213 three contributions:

$$214 \quad v^{B100} = v^{id}(1 + v_r^E + v_r^{chem}) \quad (13)$$

215

216 where v^{id} is the molar volume of the ideal solution with the exact ester profile, v_r^E is the
217 relative excess molar volume (v^E/v^{id}) and v_r^{chem} is the relative molar change caused by
218 error in chemical composition and by impurities. Because biodiesels are composed of non-
219 associated components belonging to a chemical family characterized by weak molecular
220 interactions, v_r^E is limited and the effect of pressure on v_r^E therefore has extremely little
221 impact on $v^{B100}(P)$. Furthermore, impurities lead to systematic deviations and so pressure
222 change has little impact on v_r^{chem} . Consequently, considering that the effect of pressure on v_r^E

223 and v_r^{chem} is of second order on $v^{B100}(P)$, the volume of a biodiesel at a given pressure can
224 be expressed as follows:

$$225 \quad v^{B100}(P, T) = \frac{v^{id}(P, T)}{v_{atm}^{id}(T)} v_{atm}^{B100}(T) \quad (14)$$

226

227 This equation was rewritten in terms of density in order to predict the high pressure density of
228 biodiesel based on experimental densities at atmospheric pressure $\rho_{atm}^{B100}(T)$ and on ester
229 profiles according to:

$$230 \quad \rho^{B100}(P, T) = \rho_{atm}^{B100}(T) \frac{\sum_i x_i A_i}{\sum_i x_i A_i (1 + B_i \bar{p})^{C_i}} \quad (15)$$

231

232 where x_i is the molar fraction of ester i in the biodiesel system investigated, A_i , B_i and C_i are
233 ester parameters calculated from eqs. 8, 2 and 3 respectively using the coefficients of pure
234 components listed in Table 4.

235

236

237 **4. Results and discussion**

238 Various biodiesels from different feedstocks were considered to test the predictive capacities
239 of the proposed method. They included vegetable oils such as canola (C), cottonseed, (Ct),
240 linseed (Ls), palm (P), rapeseed (R) and soybean (S), alga such as spirulina platensis (Asp),
241 animal fats such as lard (L) and tallow (T) and also waste cooking oils (W) for which high
242 pressure data are available in the literature. In addition, density data of a Purified Biodiesel
243 (PB) and biofuel blends corresponding to palm + rapeseed (PR), soybean + rapeseed (SR),
244 soybean + palm (SP), and soybean + rapeseed + palm (SRP) mixtures were considered in the
245 comparative tests. Both methyl and ethyl biodiesels were included in the database. However,
246 because there are a greater number of experimental studies on methyl esters than on ethyl

247 esters, a more in-depth analysis was conducted on FAMEs than on FAEEs. The fatty acid
248 alkyl ester profiles of each biodiesel after normalization to 100 are summarized in Table 6.
249 Eq. 15 was used to predict the densities of these biodiesels under high pressure based on their
250 fatty acid alkyl ester profiles and densities at atmospheric pressure. The statistical comparison
251 of these predictions with the data taken from the literature is summarized in Table 7 where the
252 average deviations, average absolute deviation and the maximum deviation are reported for
253 each dataset. As can be observed in Table 7, 2900 data points of 30 biodiesels were used as
254 database for model comparison. The majority of the datasets concern FAME biodiesels and
255 only four of them pertain to FAEE biodiesels. The database span a broad temperature range
256 from 278 to 413 K, and pressures of up to 200 MPa. However, most of the data are within the
257 0.1 to 60 MPa bracket. Eight datasets span pressures of more than 60 MPa, and in this
258 ensemble only three datasets reach the pressure limit imposed by the density correlation (200
259 MPa). Most of the experimental density values were obtained by direct volume change or
260 from measurements acquired using an oscillating U-tube density meter, with an additional
261 three datasets obtained using the speed of sound integration method. The expanded relative
262 uncertainty $U_r(\rho)$ reported by the authors with a level of confidence = 0.95 ($k_p = 2$) usually
263 stood at around 0.2% in the case of direct volume change measurements and at 0.1% in the
264 case of measurements obtained with the oscillating U-tube density meter. Only Pratas et al.
265 [13] reported data that are subject to uncertainties of less than 0.1%, with a claimed
266 uncertainty of 0.1 kg.m³ for the entire pressure and temperature range studied in their work.
267 Finally, the combined expanded uncertainties of density values obtained at higher pressures of
268 investigation using the speed of sound integration method were reported to be 0.2% up to 100
269 MPa and 0.3% between 100 and 200 MPa.

270

271 The comparison results in Table 7 indicate that most of the deviations are within the
272 uncertainty values of the experimental method for the FAME biodiesels, with the exception of
273 the biodiesel produced from rapeseed oil and studied by Kielczynski et al. [71]. In this case,
274 the deviation between the predictions of the correlation and the experimental data reaches 8%
275 at 200 MPa. However, for this system, the experimental change in density caused by pressure
276 change is much lower than the values of the pure C18:1 and C18:2, the main components of
277 rapeseed oil biodiesels. Similarly, calculations deviate systematically by 0.5% on average
278 with the measurements of NguyenThi et al. [75] performed on biodiesel produced from waste
279 cooking oils. This systematic bias stems from the fact that the experimental density data
280 reported in this reference are higher than the values of the main pure components (C16:0,
281 C18:0 and C18:1) of this biodiesel in the same conditions. The differences, which are
282 systematic, are probably attributable to impurities that do not belong to the FAME family.
283 The mean absolute difference between measurements reported by the other authors for FAME
284 biodiesels and the present correlation is 0.050%, which is within the experimental uncertainty
285 of 0.1-0.3% claimed by the authors. Interestingly enough, this value is of the same order of
286 magnitude as those of pure FAME (AAD=0.035%). This confirms that excess volume and
287 impurities have a second order effect on the change in density with respect to pressure and
288 therefore they can be taken into account only through density data at atmospheric pressure.
289 Moreover, the overall bias being 0.02%, the model gives no under or over estimate of the
290 pressure effect on biodiesel density.

291

292 Comparisons of experimental and predicted densities are shown in Fig. 1 for soy biodiesel, for
293 which several experimental measurements were carried out under high pressure conditions.
294 By examining this figure corresponding to $T = 313$ K, a temperature investigated in the
295 different experimental studies, it can be seen that the proposed correlation is in very good

296 agreement with the different measurements whatever the pressure, from 0.1 to 200 MPa. In
297 particular, it can be noted that the deviations are either positive or negative, which means that
298 that there is no systematic bias in the prediction method.

299 Good agreement has also been found with the results reported by Tat and Van Gerpen [23]
300 and Ivaniš et al. [72] for FAEE biodiesels (overall AD = -0.014%; overall AAD = -0.036%).
301 However, for these biodiesels the pressure range of the experimental data is much less broad
302 than for FAME biodiesels, with a maximum experimental pressure of 35 MPa for Tat and
303 Van Gerpen's data [23] and 60 MPa for the data reported by Ivaniš et al [72]. The values
304 reported by Ivaniš et al. with a standard uncertainty of $\pm 0.1\%$, deviate from eq. 15 by less than
305 0.15% with an average deviation equal to 0.008% and absolute deviation of 0.02%; the
306 majority of their results deviates from the correlation by less than 0.1 % and only two points
307 have deviations of approximately 0.14%. Fig. 2 featuring the data of Ivaniš et al. shows the
308 variation of the deviations against pressure at different temperatures. Here again, no
309 systematic bias can be observed whatever the temperature. Deviations observed between the
310 proposed method and the reported density data are within the reported experimental
311 uncertainties.

312 Besides the correlation scheme proposed to predict the effect of pressure on the densities of
313 FAME and FAEE biodiesels based on ester profiles and density data at atmospheric pressure,
314 the parameters of Table 4 could be used to predict biodiesel densities at either atmospheric or
315 high pressure based only on the ester profile using the following ideal combination rule:

$$316 \quad \rho^{B100}(P, T) = \frac{\sum_i x_i M_i}{\sum_i x_i A_i (1 + B_i \bar{p})^{C_i}} \quad (16)$$

317 with M_i the molecular weight of the fatty acid alkyl ester i . This simple approach could indeed
318 be used to calculate the density of synthetic mixtures of FAME or FAEE components, as in
319 these types of systems excess volumes are very small, but also, and above all, because
320 compositions are well known and impurity concentrations are very low. The major drawback

321 in extending the application of this simple mixing rule to biodiesels is that in real systems,
322 impurity concentrations can be significant and have a substantial influence on density values.
323 To illustrate this point, we have plotted on the same graph (Fig. 3) the deviations of the
324 experimental results from the predictions of either eq. 15 or eq. 16 for two different biodiesels
325 for which data span pressures of up to 140 MPa. The first biodiesel is the soybean oil
326 biodiesel investigated by Aitbelale et al. [73] whose chemical composition was analyzed in
327 detail by NMR and GC/MS analysis. It comprises 95.2% of FAME components, the main
328 impurities being soybean oil residues. The second biodiesel is a purified biodiesel studied by
329 Schedemann et al. [32] whose impurities are not specified but whose FAME profile, reported
330 by the authors, is normalized to 100%. Observation of Fig. 3 shows that the use of eq. 16
331 instead of eq. 15 leads to a shift in deviation of 0.4% for soybean oil biodiesel [73]
332 corresponding to the error in the prediction of density at atmospheric pressure. This shift in
333 deviation, of only 0.03% in the case of purified biodiesel [32], is mainly attributed to the
334 significant amount of impurities present in soybean oil biodiesel. It can be observed that the
335 proposed correlation scheme (eq. 14) makes it possible to ignore the real nature of the
336 impurities and to predict the effect of pressure on biodiesel density despite the presence of
337 transesterification residues in end biodiesel products.

338 Finally, according to eq.5 and eq.15, the isothermal compressibility of biodiesel can be
339 obtained from the ester profile and the parameters of Table 4 by using the following
340 combining rule:

$$341 \quad \kappa_T^{B100} = - \sum_i \phi_i \frac{B_i C_i}{1+B_i \bar{p}} \quad (17)$$

342 With ϕ_i the calculated volume fraction defined as:

$$343 \quad \phi_i = \frac{x_i A_i (1+B_i \bar{p})^{C_i}}{\sum_k x_k A_k (1+B_k \bar{p})^{C_k}} \quad (18)$$

344

345 As an example, deviations of compressibility determined by eqs. 17 and 18 using the
346 coefficients of Table 4 from literature values are shown in Fig. 4 as a function of pressure for
347 soybean oil biodiesel at $T = 313$ K. In these comparisons, the literature values were all
348 obtained from fitting the so-called Tait equation [77] and from analytically deriving it. It can
349 be observed in this figure that the deviations vary from -2% to $+2\%$ between 0.1 and 100
350 MPa and reach -4% at 200 MPa which is well within the overall uncertainty for this derivative
351 property [18]. Consequently, with the proposed procedure the accurate prediction of density
352 can be extended to the determination of its derivative with respect to pressure within the usual
353 uncertainty bracket achieved for such a property.

354

355

356 **5. Conclusion**

357

358 This article presents a computational procedure to predict the effect of pressure on biodiesel
359 density based on fatty acid alkyl ester profiles and density data at atmospheric pressure. A
360 Murnaghan equation was first proposed to correlate the densities of pure Fatty Acid Methyl
361 Ester and Fatty Acid Ethyl Ester as a function of pressure. Then, based on this equation of
362 state, and using existing high-pressure density data concerning either FAME or FAEE
363 compounds, a group-contribution method was proposed to estimate the densities of the fatty
364 acid alkyl ester components of biodiesels for which no high-pressure experimental data were
365 available. Finally, a calculation scheme based on ester profiles and density values at
366 atmospheric pressure was proposed to obtain further knowledge on density at high pressure.
367 This method yields very accurate predictions of biodiesel densities up to 200 MPa across a
368 wide range of temperatures from 280 to 400 K. The predictive character of the method was
369 demonstrated for both FAME and FAEE biodiesels, for which predictions were compared to
370 literature data. The overall mean average deviation obtained between the predictions and the
371 experimental data for a wide variety of biodiesels including vegetable oil, animal fat and
372 waste oil was less than 0.050% and the maximum deviations were of the same order of
373 magnitude as the experimental uncertainties. Due to its predictive and accurate features, the
374 method presented in this article can be considered very useful for predicting the effect of
375 pressure on biodiesel density. It can be used to fill the experimental knowledge gap that exists
376 for high pressure data. Besides extrapolating biodiesel density data to the high pressure range,
377 the group-contribution method is of interest for predicting the densities of highly
378 polyunsaturated fatty acid alkyl esters present in biodiesel fuels obtained from algal oils [78]
379 such as methyl eicosadienoate (C20:2), methyl eicosatrienoate (C20:3), methyl
380 eicosatetraenoate (C20:4), etc. for which no experimental data have yet been reported.

381

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383

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599

600 **Table 1.**
601 High pressure density data of liquid Fatty Acids Methyl Ester (FAME) and Fatty Acids Ethyl
602 Ester (FAEE).
603

604

	First author Ref	Year	Temperature range/K	Pressure range/MPa	Number of data	Technique
FAME						
Caprate	Ndiaye [20]	2012	293-393	0.1-100	120	U- Tube
C10:0	Ndiaye [20]	2012	293-323	0.1-210	241	Speed of sound
	Zarska [21]	2014	293-318	0.1-100	66	Speed of sound
	Su [22]	2018	293-323	0.1-20	30	Flowmeter
	Tat [23]	2003	293-373	0.1-35	35	PVT cell
Laurate	Pratas [13]	2011	283-333	0.1-45	84	U- Tube
	Zarska [21]	2014	293-318	0.1-100	65	Speed of sound
	Wang [24]	2016	283-363	0.1-60	117	U- Tube
	Aissa [25]	2017	293-413	0.1-60	210	U- Tube
	Habrioux [26]	2018	293-353	0.1-200	53	Speed of sound
	He [27]	2018	303-353	0.1-15	36	Flowmeter
	Pratas [13]	2011	293-333	0.1-45	70	U- Tube
Myristate	Ndiaye [28]	2013	303-393	0.1-80	90	U- Tube
	Zarska[21]	2014	293-318	0.1-90	40	Speed of sound
Palmitate	Tat [23]	2003	293-373	0.1-35	35	U- Tube
	Ndiaye [28]	2013	313-383	0.1-70	90	U- Tube
	Rasulov [29]	2018	365-442	1-20	42	piezometer
Stearate	Tat [23]	2003	293-373	0.1-35	35	PVT cell
	Pratas [13]	2011	293-333	0.1-45	70	U- Tube
C18:1	Outcalt [30]	2011	270-470	.08-50	132	U- Tube
	Ndiaye[31]	2013	293-393	0.1-100	121	U- Tube
	Ndiaye[31]	2013	293-393	0.1-210	241	Speed of sound
	Ndiaye[31]	2013	293-393	0.1-210	241	Speed of sound
Linoleate	Outcalt[30]	2011	270-370	.08-50	132	U- Tube
	Schedemann[32]	2013	278-367	.4-60	130	U- Tube
	Schedemann[32]	2013	278-337	.4-130	189	U- Tube
	Ndiaye[31]	2013	293-393	0.1-100	121	U- Tube
	Ndiaye[31]	2013	293-393	0.1-210	242	Speed of sound
FAEE						
Caprate	Ndiaye[20]	2012	293-393	0.1-100	121	U- Tube
C10:0	Ndiaye[20]	2012	293-323	0.1-210	242	Speed of sound
	Dzida [33]	2013	293-318	0.1-100	66	Speed of sound
Laurate	Dzida [34]	2013	293-318	0.1-100	66	Speed of sound
C12:0	Wang [24]	2016	283-363	0.1-60	117	U- Tube
	Aissa [25]	2017	293-413	0.1-60	210	U- Tube
	Habrioux [26]	2018	293-fr353	0.1-200	60	Speed of sound

	He [27]	2018	303-353	0.1-15	36	Flowmeter
Myristate	Ndiaye [28]	2013	303-393	0.1-100	117	U- Tube
C14:0	Dzida [34]	2013	293-318	0.1-100	56	Speed of sound
	Aissa [25]	2017	293-413	0.1-60	208	U- Tube
Oleate	Aissa [25]	2017	293-413	0.1-60	208	U- Tube
C18:1						

605
606
607

608 **Table 2.**

609 Atmospheric pressure density data of liquid Fatty Acids Methyl Ester (FAME) and Fatty
 610 Acids Ethyl Ester (FAEE).

611

	First author	Year	T/K	Nb	First author	Year	T/K	Nb	
	FAME				FAEE				
Caprate C10:0	Bonhorst [35]	1948	293-372	4	Gros [36]	1952	348	1	
	Gros [36]	1952	348	1	Shigley [47]	1955	308-368	5	
	Gouw [37]	1964	293-313	2	Smith[48]	1959	293	1	
	Ortega [38]	1990	298	1	Liew[49]	1992	288-353	10	
	Liew [39]	1992	283-353	15	Francesconi [50]	1998	293	1	
	Pratas[40]	2010	278-363	18	Ortega[51]	1999	298	1	
	Daridon [41]	2013	283-343	7	Hwu [52]	2004	293	1	
	Wang [42]	2016	298-323	6	Pratas [40]	2010	283-353	15	
	Sun [43]	2018	283-318	8	Daridon[41]	2013	283-373	10	
	Du [44]	2019	298-333	8	Wang[53]	2016	298-323	6	
	Li[45]	2019	298-323	6	Xia [54]	2018	283-318	8	
	Zhao [46]	2019	293-453	17	Li [55]	2019	293-323	7	
					Zhao[46]	2019	293-453	17	
	Laurate C12:0	Bonhorst [35]	1948	293-372	4	Gros[36]	1952	348	1
Gros[36]		1952	348	1	Shigley[47]	1955	308-368	5	
Gouw [37]		1964	293-313	2	Liew[49]	1992	288-353	11	
Ortega[56]		1990	298	1	Liau [57]	1998	293	1	
Liew [39]		1992	283-353	15	Smith[58]	1998	313	1	
Pratas[40]		2010	283-353	15	Ortega[51]	1999	298	1	
Wang [42]		2016	298-323	6	Cheng[59]	2001	293	15	
Sun [43]		2018	283-318	8	Pratas[40]	2010	283-353	15	
Li[45]		2019	293-323	7	Wang[53]	2016	298-323	6	
Zhao [46]		2019	293-433	15	Xia [54]	2018	283-318	8	
					Li [55]	2019	293-323	7	
					Zhao[46]	2019	303-443	15	
Myristate C14:0		Bonhorst [35]	1948	293-372	4	Gros[36]	1952	348	1
		Gros[36]	1952	348	1	Shigley[47]	1955	308-368	5
	Gouw [37]	1964	293-313	2	Liew[49]	1992	288-353	11	
	Ortega[56]	1990	298	1	Ortega[51]	1999	298	1	
	Liew[49]	1992	303-348	10	Pratas[40]	2010	283-353	15	
	Pratas[40]	2010	298-353	12	Daridon[41]	2013	293-373	9	
	Daridon[41]	2013	303-373	8	Li [55]	2019	293-323	7	
	Wang [42]	2016	298-323	6	Zhao[46]	2019	293-443	16	
	Sun [43]	2018	298-318	5					
	yang [60]	2018	303-333	7					
	Li[45]	2019	293-323	7					
	Zhao [46]	2019	293-413	13					
	Palmitate	Bonhorst	1948	310-372	3	Boelhouwer	1951	298-573	6

C16:0	[35]				[64]			
	Gros[36]	1952	348	1	Gros[36]	1952	348	1
	Gouw [37]	1964	313	1	Shigley[47]	1955	308-368	5
	Komoda	1970	323	1	Ortega[51]	1999	298	1
	[61]							
	Ott[62]	2008	308-338	7	Pratas[40]	2010	303-353	11
	Pratas[40]	2010	308-363	12	Yuan[63]	2019	333-547	12
	Daridon[41]	2013	313-373	7				
Yuan[63]	2019	333-547	12					
Palmitoleate C16:1	Watanabe	1960	298	1				
	[65]							
Stearate	Pratas[19]	2011	278-363	18				
	Bonhorst	1948	310-372	3	Boelhouwer	1951	323-513	5
C18:0	[35]				[64]			
	Boelhouwer	1951	323-513	5	Gros[36]	1952	348	1
	[64]							
	Gros[36]	1952	348	1	Shigley[47]	1955	308-368	5
	Gouw [37]	1964	313	1	Pratas[40]	2010	313-363	11
	Liew [49]	1992	313-353	9				
	Ott [62]	2008	318-338	5				
	Pratas[40]	2010	313-363	11				
Oleate C18:1	Knegt	1957	293	1	Candy [67]	2005	293.15	1
	[66]							
	Watanabe	1960	298	1	Pratas[40]	2010	278-363	18
	[65]							
	Gouw [37]	1964	293-313	2				
	Komoda	1970	298	1				
	[61]							
	Candy [67]	2005	293	1				
Ott [62]	2008	278-338	7					
Pratas[40]	2010	278-363	18					
Daridon	2013	283-373	10					
[41]								
Linoleate C18:2	Gouw [37]	1964	293-313	2	Smith [58]	1998	313	1
	Komoda	1970	298	1	Pratas[19]	2010	278-363	18
[61]								
Ott [62]	2008	278-338	7					
Pratas [40]	2010	278-363	18					
Daridon	2013	283-373	10					
[41]								
Linolenate C18:3	Gouw [37]	1964	293-313	2	Pratas[19]	2010	278-373	20
	Ott [62]	2008	278-338	7				
	Pratas[19]	2010	278-363	18				
Arachidate C20:0	Gouw [37]	1964	313	1	Pratas[19]	2011	318-373	12
	Pratas[19]	2011	323-373	11				
Gadoleate C20:1	Pratas[19]	2011	278-373	20				
Behenate C22:0	Pratas[19]	2011	333-373	9				
Erucate C22:1	Gouw [37]	1964	293-313	2				
	Pratas[19]	2011	278-363	18				
Lignocerate C24:0	Pratas[19]	2011	338-373	8				

612 **Table 3.**

613 Group contribution values for parameters of eqs. 2-3, 6-8 where the units are K for the
614 temperature T , MPa for pressure p and $\text{cm}^3 \cdot \text{mol}^{-1}$ for volume increments $v_j(T, p)$.

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	a_0	$a_1 \times 10^3$	$a_2 \times 10^6$	$b_0 \times 10^3$	$b_1 \times 10^6$	$b_2 \times 10^9$	$c_0 \times 10^3$	$c_1 \times 10^6$
CH3	16.43000	55.62000	0	141.0915	-937.170	1786.650	-18.02781	-416.5600
CH2	12.04000	14.10000	0	1.828686	-7.32110	8.242550	673.4669	-5335.580
CH=CH	43.17037	-99.28637	155.2190	4.549329	-31.8870	59.77000	-2910.446	5352.666
CH ₃ COO	45.86310	-3.709090	83.30000	44.79574	-151.000	165.7020	198.9688	-910.8600
C ₂ H ₅ COO	68.40212	-40.28484	159.8240	80.85027	-350.450	428.5890	249.4559	-1084.450

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620 **Table 4**

621 Parameters of Murnaghan equation [17] of pure Fatty Acid Methyl Esters and Fatty Acid
 622 Ethyl Esters used in eq. 15 where the units are K for the temperature T , MPa for pressure p
 623 and $\text{cm}^3 \cdot \text{mol}^{-1}$ for volume $v(T, p)$.
 624

	a_0	$a_1 \times 10^3$	$a_2 \times 10^6$	$b_0 \times 10^3$	$b_1 \times 10^6$	$b_2 \times 10^9$	$c_0 \times 10^3$	$c_1 \times 10^6$
FAME								
C10:0	167.8959	106.7739	166.9610	20.75502	-131.7700	297.3830	-97.64292	-15.01000
C12:0	198.5288	103.4472	206.0360	13.35048	-87.15400	232.0300	-101.6336	15.80980
C14:0	221.4977	143.5773	184.2740	17.60627	-119.0600	297.7900	-105.9907	57.67670
C16:0	253.8070	124.4893	255.0380	24.99391	-166.2200	360.1700	-129.2231	117.9640
C16:1	246.7464	152.5615	202.8800	8.527390	-52.22200	142.8670	-127.4771	28.27370
C18:0	258.6963	273.2155	69.19890	8.132622	-45.43100	123.5910	-135.3409	73.08580
C18:1	280.9855	117.4477	276.2360	14.03595	-78.61400	184.0230	-88.57095	-41.54900
C18:2	274.0727	123.8887	255.2240	11.17410	-54.61100	139.6520	-66.27109	-98.65500
C18:3	302.2871	-95.36963	585.8010	10.26528	-70.31300	170.1990	-172.6542	85.72460
C20:0	270.5583	375.1460	-40.78100	7.517432	-40.84400	110.5550	-145.1129	92.86160
C20:1	313.5710	95.65575	346.5100	7.681347	-45.04300	119.4270	-150.9487	83.34940
C22:0	277.7420	485.0573	-136.9800	6.946821	-36.68300	99.12680	-154.1908	110.1740
C22:1	323.6248	222.8248	186.2350	7.186587	-41.16200	108.5260	-160.8404	104.4310
C24:0	313.6635	464.3115	-88.70100	6.425814	-32.95100	89.11600	-162.6810	125.4520
FAEE								
C10:0	180.6837	124.4817	170.1120	4.639978	-17.95200	104.5670	-54.81752	-141.3800
C12:0	212.4059	115.9065	214.9730	4.976450	-32.75200	145.0720	-92.82938	-15.31200
C14:0	242.3230	115.3258	252.1790	12.42137	-66.50300	172.5440	-70.87564	-86.47500
C16:0	264.2868	162.0671	218.6960	4.164460	-15.18400	75.66210	-93.48324	-45.96500
C16:1	272.4825	85.24879	315.0430	3.146145	-10.00500	66.60260	-83.06140	-103.8100
C18:0	282.5838	228.2740	159.7660	4.292851	-16.17200	71.93060	-104.8864	-19.66100
C18:1	292.5633	139.9768	272.0640	7.814890	-52.22200	162.9790	-111.4736	27.66500
C18:2	285.3669	148.0623	242.2020	2.697067	-8.920400	60.16880	-87.92858	-122.5800
C18:3	277.2908	162.6322	211.0990	1.626377	-2.126800	47.58790	-70.13632	-203.6600
C20:0	299.6463	302.3346	86.89330	4.312686	-16.39100	67.72530	-115.2614	3.007290
C20:1	320.6425	141.6488	315.0430	3.820384	-15.23000	66.75190	-111.2576	-33.13800
C22:0	325.6321	297.3352	159.8240	4.316140	-16.60800	64.30960	-125.5823	24.99000
C22:1	344.7225	169.8488	315.0430	3.930681	-16.05400	64.50450	-122.7918	-6.357800
C24:0	349.7121	325.5352	159.8240	4.240813	-16.22100	60.35050	-134.8461	43.56520

625

626 **Table 5.**

627 Statistical characteristics (Average Deviations, Average Absolute Deviations, Maximum
 628 Deviation) for the predictions of the effect of pressure ρ/ρ_{atm} on FAME and FAEE pure
 629 components from eqs. 1-3 using parameters given in Table 4.

630

	First author	AD %	AAD %	MD %	First author	AD %	AAD %	MD %
FAME					FAEE			
Caprate	Ndiaye ¹ [20]	0.02	0.04	0.17	Ndiaye ¹ [20]	0.01	0.05	0.14
C10:0	Ndiaye ² [20]	0.004	0.03	0.10	Ndiaye ² [20]	0.004	0.02	0.07
	Zarska [21]	-0.03	0.03	0.06	Dzida [33]	-0.02	0.02	0.07
	Su [22]	0.10	0.12	0.56				
Laurate	Tat [23]	-0.01	0.02	0.06	Dzida [34]	-0.03	0.03	0.08
C12:0	Pratas [13]	0.001	0.01	0.03	Wang [24]	0.01	0.03	0.10
	Zarska [21]	-0.01	0.05	0.25	Aissa [25]	0.004	0.03	0.21
	Wang [24]	-0.02	0.02	0.21	Habrioux [26]	-0.03	0.05	0.23
	Aissa [25]	0.00	0.02	0.16				
	Habrioux [26]	-0.04	0.06	0.30				
	He [27]	0.02	0.02	0.08				
Myristate	Pratas [13]	0.02	0.02	0.05	Ndiaye [28]	0.00	0.03	0.12
C14:0	Ndiaye [28]	-0.03	0.09	0.40	Dzida [34]	-0.02	0.02	0.06
	Zarska [21]	-0.03	0.03	0.11	Aissa [25]	0.01	0.02	0.08
Palmitate	Tat [23]	-0.08	0.08	0.16				
C16:0	Ndiaye [28]	-0.07	0.07	0.33				
	Rasulov [29]	-0.14	0.15	0.60				
Stearate C18:0	Tat [23]	-0.24	0.24	0.71				
Oleate C18:1	Pratas [13]	-0.01	0.01	0.03				
	Outcalt [30]	-0.01	0.02	0.10				
	Ndiaye ¹ [31]	0.01	0.03	0.07				
	Ndiaye ² [31]	-0.01	0.02	0.05				
Linoleate C18:2	Outcalt [30]	-0.01	0.02	0.12				
	Schedemann [32]	-0.01	0.02	0.05				
	Schedemann [32]	-0.02	0.02	0.08				
	Ndiaye ¹ [31]	0.02	0.03	0.14				
	Ndiaye ² [31]	0.01	0.02	0.08				

631

632 ¹ obtained from U-tube vibrating densimeter

633 ² obtained from speed of sound integration

634

635

636 **Table 6.**

637 Fatty Acid Alkyl Ester profiles in mol% of different oil and fat biodiesels used for testing the
 638 predictive capacity of the proposed method.

639

Biodiesel* / Author	C1 0:0	C1 2:0	C1 4:0	C1 6:0	C1 6:1	C1 8:0	C1 8:1	C1 8:2	C1 8:3	C2 0:0	C2 0:1	C2 2:0	C2 2:1	C2 4:0
Methyl														
C Tat [23]	0.0	0.0	0.0	4.3	0.2	1.9	65.0	19.1	9.5	0.0	0.0	0.0	0.0	0.0
Ct Prieto [14]	0.0	0.0	1.1	28.4	0.0	2.7	17.3	50.5	0.0	0.0	0.0	0.0	0.0	0.0
Ls1 Tat [23]	0.0	0.0	0.0	22.4	0.0	27.4	15.0	7.5	27.7	0.0	0.0	0.0	0.0	0.0
Ls2 Tat [23]	0.0	0.0	0.0	40.9	0.0	42.3	4.6	2.3	9.9	0.0	0.0	0.0	0.0	0.0
P Pratas [13]	0.0	0.3	0.7	44.6	0.1	3.8	40.3	9.5	0.1	0.3	0.1	0.1	0.0	0.1
R Pratas [13]	0.0	0.1	0.1	5.7	0.2	1.6	61.8	21.1	7.0	0.5	1.2	0.3	0.2	0.2
R Habrioux [70]	0.0	0.1	0.1	5.7	0.2	1.6	61.8	21.1	7.0	0.5	1.2	0.3	0.2	0.2
R Kielczynski [71]	0.0	0.0	0.1	5.0	0.4	1.8	60.6	20.0	9.2	0.5	1.5	0.2	0.4	0.3
S Tat [23]	0.0	0.0	0.1	11.9	0.1	3.9	23.1	53.1	7.8	0.0	0.0	0.0	0.0	0.0
S Pratas [13]	0.0	0.0	0.1	11.6	0.1	3.9	22.7	53.2	7.0	0.3	0.2	0.7	0.2	0.0
S Habrioux [70]	0.0	0.0	0.1	11.6	0.1	3.9	22.7	53.2	7.0	0.3	0.2	0.7	0.2	0.0
S Ivaniš [72]	0.0	0.0	0.1	8.7	0.0	3.2	28.9	58.9	0.0	0.0	0.0	0.0	0.2	0.0
S Aitbelale [73]	0.0	0.0	0.0	12.1	0.0	4.0	23.6	53.2	7.1	0.0	0.0	0.0	0.0	0.0
S ^H Tat [23]	0.0	0.0	0.0	12.3	0.0	87.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
S ^O Tat [23]	0.0	0.0	0.0	16.4	0.0	5.2	56.1	22.3	0.0	0.0	0.0	0.0	0.0	0.0
Asp Bessières [74]	0.0	0.0	5.3	45.7	9.2	2.5	1.7	21.8	13.8	0.0	0.0	0.0	0.0	0.0
L Tat [23]	0.0	0.1	1.8	27.2	3.4	12.2	45.2	10.0	0.1	0.0	0.0	0.0	0.0	0.0
L Ivaniš [72]	0.0	0.0	0.0	30.5	0.0	16.4	42.3	10.4	0.2	0.0	0.0	0.0	0.2	0.0
T Tat [23]	0.0	0.1	4.1	28.4	3.8	21.7	39.1	2.2	0.6	0.0	0.0	0.0	0.0	0.0
W NguyenThi [75]	0.0	0.0	3.8	26.9	7.7	12.8	36.7	9.6	0.0	0.5	2.0	0.0	0.0	0.0
W Aitbelale [76]	0.0	0.0	67.4	0.0	0.0	0.0	24.5	7.4	0.7	0.0	0.0	0.0	0.0	0.0
BP Schedemann [32]	0.0	0.0	0.6	8.6	0.2	2.2	58.1	20.5	8.1	0.4	1.0	0.1	0.1	0.1
RP Pratas [13]	0.0	0.3	0.6	24.7	0.2	2.9	51.8	15.2	3.0	0.4	0.6	0.2	0.1	0.0
SP Pratas [13]	0.0	0.2	0.0	27.3	0.1	3.9	32.3	31.3	3.5	0.3	0.2	0.3	0.1	0.5
SR Pratas [13]	0.0	0.0	0.1	9.7	0.2	2.7	41.4	37.4	7.0	0.4	0.6	0.4	0.1	0.0
SRP Pratas [13]	0.0	0.2	0.5	20.4	0.2	3.2	41.6	27.6	4.6	0.4	0.5	0.3	0.1	0.4
Ethyl														
S Tat [23]	0.0	0.0	0.0	15.1	0.0	8.8	52.0	24.1	0.0	0.0	0.0	0.0	0.0	0.0
S Ivaniš [72]	0.0	0.0	0.1	8.7	0.0	3.2	29.0	59.0	0.0	0.0	0.0	0.0	0.0	0.0
S ^H Tat [23]	0.0	0.0	0.0	12.9	0.0	87.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
S ^O Tat [23]	0.0	0.0	0.1	11.9	0.1	3.9	23.2	53.0	7.8	0.0	0.0	0.0	0.0	0.0

640 * C: canola, Ct: cottonseed, Ls: linseed, P: palm, R: rapeseed, S: soybean, Asp: alga spirulina

641 platensis, L: lard, T: tallow, W: waste cooking oils, PB: purified biodiesel, PR: palm + rapeseed

642 blend, SR: soybean + rapeseed blend, SP: soybean + palm blend, SRP: soybean + rapeseed + palm

643 blend, ^H Hydrogenated, ^O : Oxidized.

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645 **Table 7.**

646 Statistical Characteristics (Average Deviations, Average Absolute Deviations, Maximum
 647 Deviation) for the prediction of the effect of pressure on biodiesel densities from eq. 15 using
 648 parameters given in Table 4.

649

Biodiesel* / First author	Year	AD	AAD	MD	T range /K	P range /MPa	Ur(ρ) %	Number of data
Methyl								
C Tat [23]	2003	0.01	0.03	0.09	293-373	0.1-35	0.2	30
Ct Prieto [14]	2015	0.01	0.02	0.11	288-358	0.1-30	0.17	120
Ls1 Tat [23]	2003	-0.01	0.03	0.12	293-373	34.5	0.2	30
Ls2 Tat [23]	2003	-0.05	0.06	0.26	293-373	34.5	0.2	30
P Pratas [13]	2011	0.007	0.01	0.06	283-333	0.1-45	0.02	84
R Pratas [13]	2011	0.005	0.01	0.05	283-333	0.1-45	0.02	84
R Habrioux [70]	2013	0.14	0.14	0.28	293-393	0.1-200	0.3	231
R Kielczynski [71]	2017	4.42	4.42	8.05	278-293	0.1-250	0.2	104
S Tat [23]	2003	0.01	0.03	0.09	293-373	0.1-35	0.2	30
S Pratas [13]	2011	0.02	0.02	0.07	283-333	0.1-45	0.02	84
S Habrioux [70]	2013	0.03	0.05	0.13	293-393	0.1-200	0.3	231
S Ivaniš [72]	2016	0.03	0.03	0.12	288-413	0.1-60	0.1	222
S Aitbelale [73]	2019	0.04	0.05	0.14	298-393	0.1-140	0.1	136
S ^H Tat [23]	2003	-0.13	0.13	0.38	293-373	0.1-35	0.2	30
S ^O Tat [23]	2003	0.03	0.04	0.10	293-373	0.1-35	0.2	30
Asp Bessières [74]	2018	0.01	0.02	0.05	293-353	0.1-140	0.1	51
L Tat [23]	2003	-0.01	0.04	0.12	293-373	0.1-35	0.2	30
L Ivaniš [72]	2016	-0.04	0.04	0.18	298-413	0.1-60	0.1	194
T Tat[23]	2003	-0.05	0.06	0.22	293-373	0.1-35	0.2	30
W NguyenThi [75]	2018	0.46	0.46	0.94	293-353	0.1-140	0.1	60
W Aitbelale [76]	2019	0.20	0.20	0.39	298-393	0.1-140	0.1	136
BP Schedemann [32]	2013	-0.02	0.03	0.06	288-397	0.1-130	0.1	324
RP Pratas [13]	2011	0.004	0.006	0.02	283-333	0.1-45	0.02	84
SP Pratas [13]	2011	0.003	0.01	0.04	283-333	0.1-45	0.02	84
SR Pratas [13]	2011	0.02	0.02	0.06	283-333	0.1-45	0.02	84
SRP Pratas [13]	2011	0.001	0.004	0.01	283-333	0.1-45	0.02	84
Ethyl								
S Tat [23]	2003	-0.08	0.08	0.25	293-373	0.1-35	0.2	30
S Ivaniš [72]	2016	0.008	0.02	0.14	288-373	0.1-60	0.1	196
S ^H Tat [23]	2003	-0.1	0.1	0.4	293-373	0.1-35	0.2	30
S ^O Tat [23]	2003	-0.004	0.03	0.09	293-373	0.1-35	0.2	30

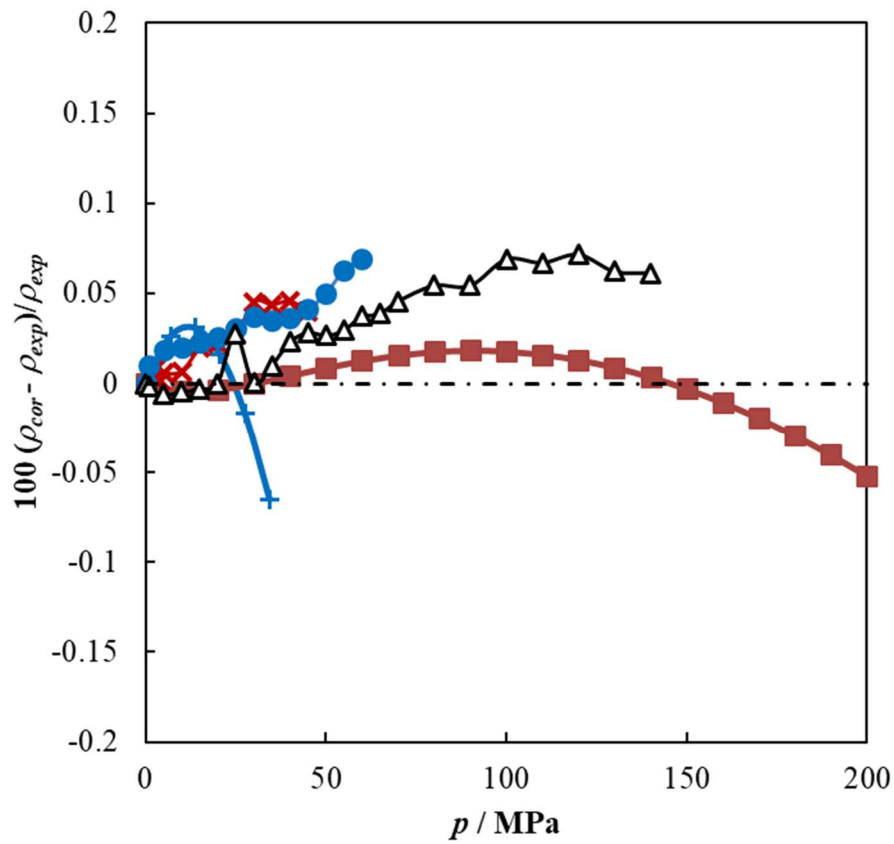
650 * C: canola, Ct: cottonseed, Ls: linseed, P: palm, R: rapeseed, S: soybean, Asp: alga spirulina
 651 platensis, L: lard, T: tallow, W: waste cooking oils, PB: purified biodiesel, PR: palm +
 652 rapeseed blend, SR: soybean + rapeseed blend, SP: soybean + palm blend, SRP: soybean +
 653 rapeseed + palm blend, ^H Hydrogenated, ^O : Oxidized.

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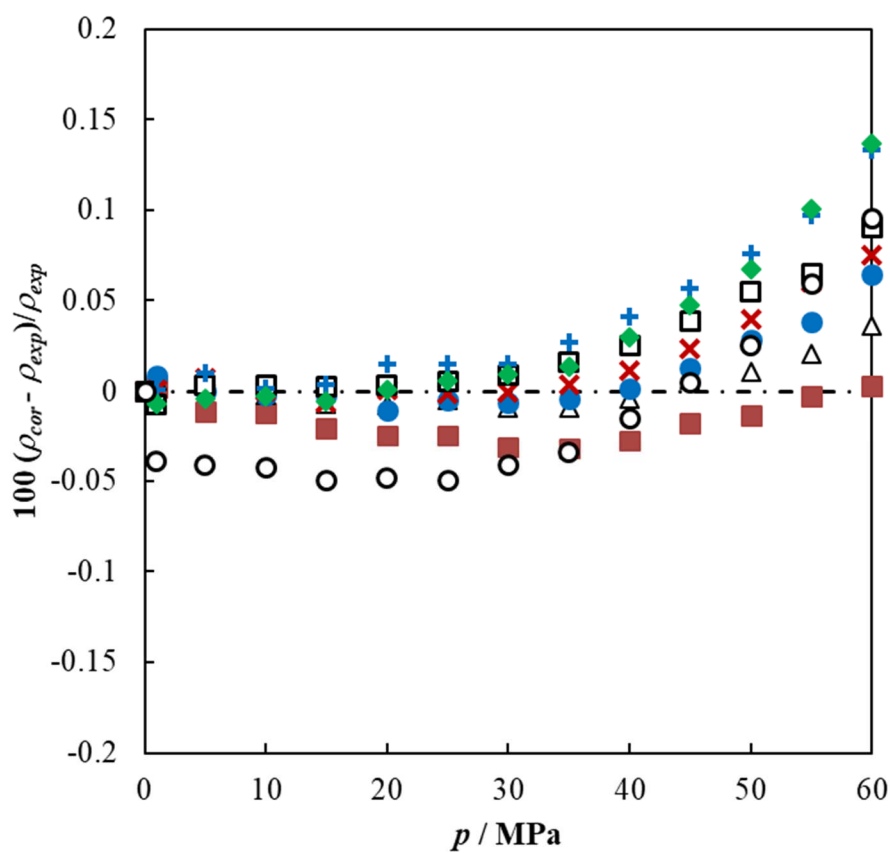
Fig. 1. Relative density deviations between the calculated density values with the proposed method (eq. 15) and the experimental literature values for soy bean oil methyl biodiesel at $T = 313$ K: +, Tat and Van Gerpen [23]; ×, Pratas et al. [13]; ■, Habrioux et al. [70]; ●, Ivaniš et al. [72]; Δ, Aitbelale et al. [73].

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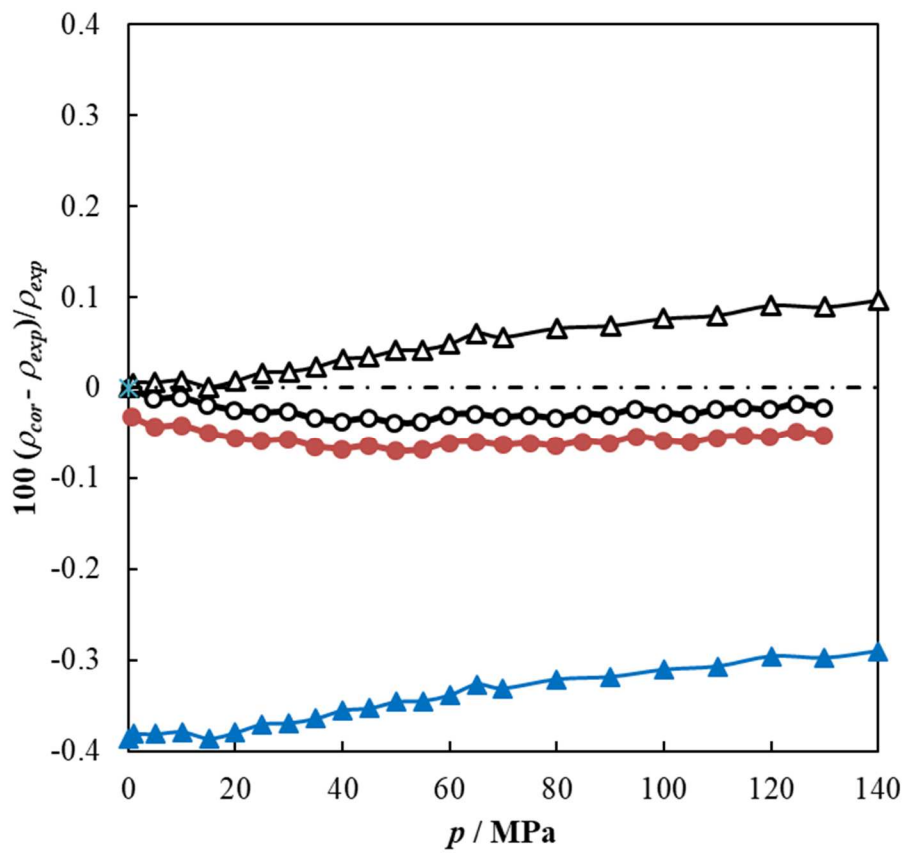
Fig. 2. Relative density deviations between the calculated values with the proposed method (eq. 15) and the experimental literature values of vaniš et al. [72] for soy bean oil ethyl biodiesel at different temperatures: ■, 293 K ; Δ, 303 K; ●, 313 K; ×, 323 K; □, 333 K; +, 353 K; ◆, 363 K; ○, 373 K



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711 **Fig. 3.** Comparison of the density deviations obtained from eqs. 15 and 16 for two methyl
 712 biodiesels at $T=298$ K: Δ , soy bean oil biodiesel [73] with eq. 15; \blacktriangle soy bean oil biodiesel
 713 [73] with eq. 16: \bullet , purified biodiesel [32] with eq. 15; \bullet , purified biodiesel [32] with eq.
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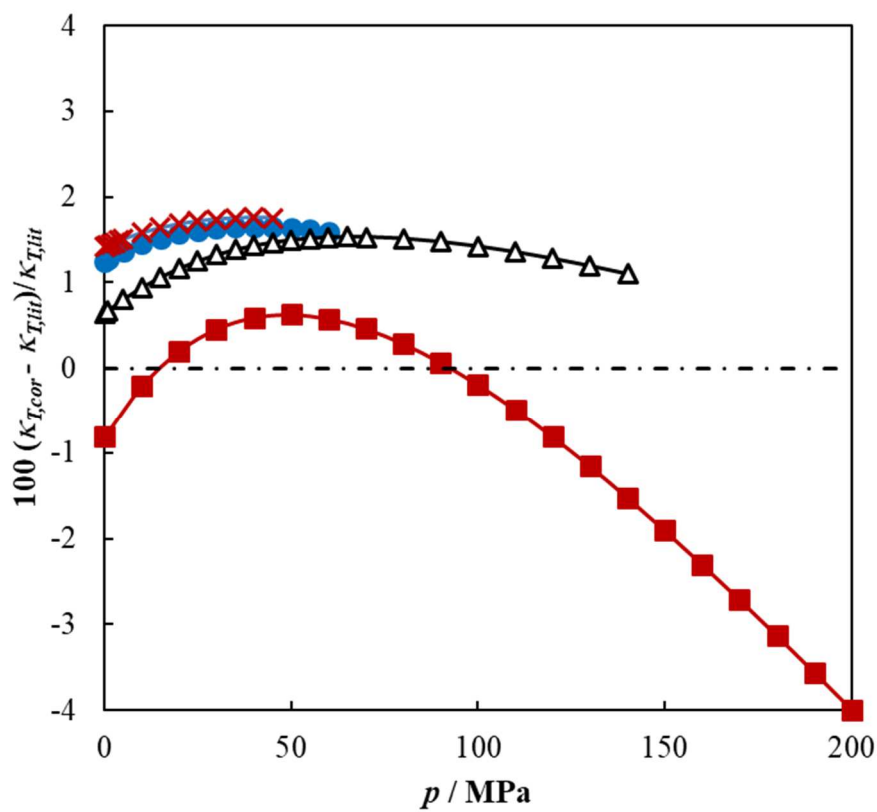
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723 **Fig. 4.** Relative compressibility deviations between the calculated values with the proposed
 724 method (eqs. 17, 18) and the literature values obtained from fitting a Tait like equation of
 725 state for soy bean oil methyl biodiesel at $T = 313$ K: \times , Pratas et al. [13]; \blacksquare , Habrioux et al.
 726 [70]; \bullet , Ivaniš et al. [72]; Δ , Aitbelale et al. [73].

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