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Temperature Dependence of Density, Viscosity, Thermal Conductivity and Heat Capacity of Vegetable Oils for Their Use as Biofuel in Internal Combustion Engines

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Abstract

This work gives tools to overcome the difficulty to determine experimentally physical properties for vegetable oils within the range of temperature typically observed during the injection phase in a diesel engine. Knowing vegetable oils' physical properties to these ranges of temperature is of fundamental importance when modeling their combustion in diesel engine. However, vegetable oils' experimental physical properties data are rare in the literature for temperature above 523 K. This paper describes experimental measurements and estimation methods for density, dynamic viscosity, thermal conductivity and heat capacity of vegetable oils for this particular range of temperature. The methodology uses several correlative methods using group contribution approach for each property and compares experimental data with predicted one to select the more accurate model. This work has shown the rapeseed and jatropha oils' physical properties can be satisfactorily predicted as a function of temperature using group contribution approach.

Keywords

Rapeseed Oil, Jatropha Oil, Physical Properties, Group Contribution Method, Energy, Diesel Engine

1. Introduction

In the last three decades, several studies have shown the potential of pure vege-

table oils as fuel in diesel engines and burners [1] [2] [3] [4] [5]. Their applications are numerous for developing countries: agriculture, irrigation, power and heat generation as well as drinking water supply. However, there are still problems related to their chemical and physical properties such as fatty acid composition, viscosity and volatility [6] [7] [8] [9] [10]. Their use may lead to the formation of carbon deposits under certain temperature conditions. These deposits can lead rapidly to engine mechanical failure. Studies have been conducted to determine the causes and possible solutions to the problems encountered in using vegetable oils as fuel in diesel engines [6] [11].

Many studies, mostly experimental, have described the mechanisms of evaporation and combustion of vegetable oils under different conditions of temperature and pressure [12] [13] and have determined their characteristics of evaporation. Attempts of models to calculate these characteristics are facing difficulties to correctly describe the transient phase. This phase of heating and dilatation takes place before the vaporization of vegetable oils droplets when they are introduced in diesel engines combustion chamber which minimum working temperature is above 450 K. Taking account of this transient phase requires the knowledge of the physical properties such as density, viscosity, thermal conductivity and specific heat capacity, which data are rare in literature for temperature above 450 K. Density, thermal conductivity, dynamic viscosity and heat capacity are useful for selecting fuels for internal combustion engines and have normative values in quality standards for commercial fuels used in compression ignition engines. Recent researches on the determination of the physical properties of alternative fuels have focused on biodiesel [14] [15] [16] [17] [18] or blends between diesel and other substances or mixtures [19] [20] [21] [22] [23]. Only a few studies have been interested in pure vegetable oils [24] [25] [26]. Indeed, determining vegetable oils physical properties is difficult because their composition varies from one oil to another. Furthermore, experimental measurements of these properties are difficult or impossible to perform over a certain level of temperature because of their decomposition [27]. Methods such Static Method and a Flow Method [28], Low Residence Time Flow Method [29] and the Pulse-Heating Method [30] are much more used to determine critical properties of pure compounds or unstable substances and mixtures.

Vegetable oils are mainly used for food purposes and therefore there are few data in literature on their physical properties up to 450 K. This lack of data had to be overcome by experimental measurements and predictions for industrial use of vegetable oils. The main objective of this work is to determine experimentally vegetable oils density, viscosity, thermal conductivity and heat capacity up to 523 K and hence to predict the same physical properties for temperature above 523 K by means of predictive methods based on group contribution approach. At least two group contribution methods were tested for rapeseed and *Jatropha* oil density, viscosity, thermal conductivity and heat capacity which can be obtained experimentally or available in the literature. The physical properties

above mentioned are evaluated as a function of temperature and the models which give very good agreement with literature or experimental data obtained in this work will be retained. The contribution of this work is to provide rapeseed and *Jatropha* experimental physical properties and predictive one by group contribution methodology that can be applied to others vegetable oils. Group contribution method has been used in recently works to estimate vegetable oils density [25] and viscosity [31], lower heating value [32], cetane number of biodiesel fuel [33], and fatty acid compounds viscosities [15], but few of them has determined properties above 450 K. *Jatropha curcas* and rapeseed oils were chosen because they are respectively representative of southern and northern hemisphere first generation biofuels and also because of the availability of their physical properties data in the literature.

The critical properties such as critical temperature, critical pressure or critical volume are very important because they are involved in determining the physical properties mentioned above. Then, the critical properties and normal boiling point will be firstly determined by correlative method.

2. Materials and Methods

Experimental tests were carried out at CIRAD Biomass Energy Laboratory (UR BioWooEB) in Montpellier (France), with the collaboration of the “Laboratoire de Physique et de Chimie de l’Environnement” (Burkina Faso), and the PROMES-CNRS laboratory of Perpignan (France).

2.1. Vegetable Oils

Jatropha curcas oil was obtained from agricultural producers in Burkina Faso. Rapeseed oil is commercially available and was purchased in a refined standard state from a food reseller in France.

The physical and chemical characteristics in standard conditions of *jatropha curcas* and rapeseed oils used in this study were determined at BioWooEB. They are listed in **Table 1**. The density values were obtained using a pycnometer following the NF EN ISO 12185 procedure. The dynamic viscosities were measured according to the NF EN ISO 3104. The flashpoints were obtained with a Pensky-Martens apparatus and using the NF EN ISO 2719 normative. We obtained the surface tension values by using the NF ISO 6295. Whereas lower heating values were measured in an Anton Paar Calorimeter type 6200 according to the NF EN 14918 method for solid biofuels, adapted and applied to liquids. The

Table 1. Physical characteristics of rapeseed oil and *jatropha curcas* oil.

| | Density (kg/m ³ at 288 K) | Kinematic viscosity (mm ² /s at 313 K) | Flash point (K) | Surface tension (N/m) | Low heating value (MJ/kg) | Carbon residue (%) |
|---------------------|--|---|--------------------|--------------------------|------------------------------|-----------------------|
| Rapeseed oil | 925 | 34.9 | 483 | 32.9 | 37.1 | 0.39 |
| <i>Jatropha</i> oil | 940 | 34 | 498 | - | 36.3 | - |

carbon residue was measured by performing the NF EN ISO 10370 operation in a furnace of 773 K. Some of these physical properties were determined at the IESPM on the request of the unit BioWooEB.

The fatty acid composition was determined by gas chromatography, using a Agilent 6890 GC type with a FID detector and a CP-WAX 58CB column (25 m × 0.32 mm × 0.2 μm). The fatty acid composition of the oils is reported in **Table 2**.

2.2. Methods

2.2.1. Experimental

In this section the experimental methods used to characterize the considered oils in the 298 - 523 K temperature range are described. The different methods or devices used in this work for the determination of density, dynamic viscosity, thermal conductivity and heat capacity of rapeseed and jatropha oils are listed in **Table 3**. These physical properties were carried out at the PROMES-CNRS laboratory in Perpignan.

The details of method 3 ω can be obtained in the following literature [34].

2.2.2. Correlative Methods

A model that could predict pure vegetable oils physical properties based on the knowledge of their fatty acid composition would be useful in their direct use as fuel or in the optimization of biodiesel production processes or for the blending with others suitable products. On the basis of the fatty acid composition of the vegetable oils, the group contribution methods are known to be a powerful tool for predicting physical properties when experimental data are not available [15] [35]. Indeed, more than 95% of vegetable oil composition includes fatty acids. Furthermore, group contribution method is very effective to take into account of the contribution of glycerol that cannot be neglected for a more accurate estimate.

Table 2. Measured Fatty acids composition (peak area, %) of rapeseed and jatropha oils.

| Fatty acids | Formula | Rapeseed oil | Jatropha oil |
|-------------------------|--|--------------|--------------|
| Oleic (C18:1) | C ₁₈ H ₃₄ O ₂ | 60.78 | 41.64 |
| Linoleic (C18:2) | C ₁₈ H ₃₂ O ₂ | 19.22 | 32.53 |
| Linolenic (C18:3) | C ₁₈ H ₃₀ O ₂ | 8.92 | 0.00 |
| Palmitic (C16:0) | C ₁₆ H ₃₂ O ₂ | 4.78 | 16.00 |
| Stearic (C18:0) | C ₁₈ H ₃₈ O ₂ | 1.35 | 6.05 |
| Other minor fatty acids | - | 4.95 | 3.77 |

Table 3. Experimental devices or methods for rapeseed and jatropha oils physical properties determination.

| Physical Properties | Density | Dynamic viscosity | Thermal conductivity | Heat capacity |
|---------------------|------------|-------------------|----------------------|-----------------|
| Device/Method | Pycnometer | Rheometer ARES-G2 | 3 ω Method | DSC Setaram C80 |

In this study, rapeseed and jatropha oils critical properties were estimated by using correlative methods that are based on group-contribution approach. Marrero and Gani (MG) method was used [36]. In fact, several studies [26] [36] [37] [38] show that this method performs better in terms of greater accuracy and wide applicability. Marrero and Gani method performs estimations at different levels: the primary level describes a wide variety of simple, monofunctional groups; the higher levels treat multifunctional structures and take into account the interactions among isomers functionalities.

For this method, each critical property is estimated by a function f which depends, on the one hand on the different contributions of the functional groups at different levels as shown in the Equation (1) and on the other hand on the primary properties. The functions used for this work are listed in **Table 4**.

$$f(X) = \sum_i N_i C_i + w \sum_j M_j D_j + z \sum_k O_k E_k \quad (1)$$

$f(X)$ is a function of the property X to be estimated, and i , j and, k refer to the first, second and third order groups defined in the group contribution method. N_i and M_j are the number of the i -th first order group, and the j -th second order group, respectively, present in the molecule, and C and D are the fitted contributions to the first and second order groups, respectively.

The approach used is based on the rapeseed and jatropha oils fatty acid composition: each fatty acid has been fragmented into several chemical groups and the contribution of each group is taken into account to get the contribution of the corresponding fatty acid. Then, the rapeseed or Jatropha oils critical properties can be estimated satisfactorily by taking into account their composition in fatty acids. For these different physical properties, at least two methods most suited for vegetable oils were considered and the best of them was retained.

Density estimation

There are several methods that can be used to estimate liquid density of pure or mixture compound [39]. However, the most important and accurate among them, and that is applicable to vegetable oils is Gunn Yamada's and Method of Ihmels *et al.* [38].

According to Gunn Yamada's estimation method, the pure compound liquid density is evaluated as follows Equations (2) to (6). The temperature range of this correlation extends from a reduced temperature of 0.20 to just below the critical temperature.

Table 4. Functions and constants for Marrero and Gani's group estimation method.

| Properties | Function f | Constants |
|----------------------|--|---|
| Boiling Point | $f(X) = \exp(T_b/T_{bo})$ | $T_{bo} = 222.543 \text{ K}$ |
| Critical Temperature | $f(X) = \exp(T_c/T_{co})$ | $T_{co} = 231.239 \text{ K}$ |
| Critical Pressure | $f(X) = (P_c - P_{c1})^{0.5} - P_{c2}$ | $P_{c1} = 5.9827 \text{ bar}, P_{c2} = 0.108998 \text{ bar}^{-0.5}$ |
| Critical volume | $f(X) = V_c - V_{co}$ | $V_{co} = 7.95 \text{ cm}^3/\text{mol}$ |

$$\frac{1}{\rho} = V_{sc} V_R^{(o)} (1 - \omega \Gamma) \quad (2)$$

where

$$V_{sc} = \frac{1}{\frac{\rho_{ref}}{V_R^{(o)}(T_{ref}) [1 - \omega \Gamma(T_{ref})]}} \quad (3)$$

$$\text{and } \Gamma = 0.29607 - 0.09045T_R - 0.04843T_R^2 \quad (4)$$

and ω is the acentric factor and is calculated using Equation (5)

$$\frac{P_c V_c}{RT_c} = 0.291 - 0.080\omega \quad (5)$$

where P_c , T_c and V_c are the critical pressure, temperature and volume, respectively. T_{ref} is a reference temperature, generally ambient temperature, ρ_{ref} is the density at the reference temperature.

$$V_R^{(o)} = 0.33593 - 0.33593T_R + 1.51941T_R^2 - 2.02512T_R^3 + 1.11422T_R^4 \quad (6)$$

for temperature ranges corresponding to $0.2 \leq T_R \leq 0.8$ where T_R is the reduced temperature with $T_R = \frac{T}{T_c}$. In this study, reference data are these obtained experimentally at 298 K from this study.

Ihmels and Gmehling [38] extended and revised the group contribution method GCVOL developed by Elbro *et al.* for the prediction of liquid density. According to this method, the density of vegetable oils can be estimated by the Equation (7) below:

$$\rho = \frac{M_w}{V} = \frac{M_w}{\sum n_i \Delta v_i} \quad (7)$$

where M_w is the molecular weight and V the molar volume. The molar volume is obtained by summing up all the group volume contributions Δv_i with n_i the number of group i appearing in the compound, while Δv_i is expressed as a polynomial function of absolute temperature:

$$\Delta v_i = A_i + B_i T + C_i T^2 \quad (8)$$

where the units are K for temperature and $\text{cm}^3 \cdot \text{mol}^{-1}$ for Δv_i . Group functional and there parameters A_i , B_i and C_i can be seen in this literature [39].

The molecular weight of vegetable oils can be estimated using Equation (9):

$$M_w = 3 \sum x_i M_{wi} + 38.0488 \quad [40] \quad (9)$$

Dynamic viscosity estimation

The most important methods used for dynamic viscosity estimation of pure compounds are based on group contribution models proposed by Jöback-Lydersen's [39] and by Morris [41].

Jöback-Lydersen's method used a simple correlation given by Equation (10):

$$\mu = M_w \cdot \exp \left[\frac{\sum \Delta \mu_a - 597.82}{T} + \sum \Delta \mu_b - 4.294 \right] \quad [42] \quad (10)$$

where $\Delta\mu_a$ and $\Delta\mu_b$ are Jöback groups' contributions which are given and T is the temperature. No temperature limitations are specified for this method except for the fact that the temperature must be less than the critical.

For Morris's Method, the dynamic viscosity μ_L can be estimated using Equation (11) to Equation (12).

$$\log_{10} \frac{\mu_L}{\mu^+} = J \left(\frac{1}{T_R} - 1 \right) \quad (11)$$

$$\text{where } J = (0.577 + \sum \Delta\mu_M)^{1/2} \quad (12)$$

$\Delta\mu_M$ represents the group contributions factors which are given and μ^+ is compound class group contribution. This method is limited to temperatures less than 0.8 times the critical temperature.

Thermal conductivity estimation

For thermal conductivity estimation of rapeseed and jatropha oils respectively, two methods based on group contribution method proposed by Sastri, and Sato-Riedel [39] were selected. Sastri method gives the thermal conductivity λ_L by Equation (13) below:

$$\lambda_L = \lambda_b a^m \quad (13)$$

where m is given by Equation (14)

$$m = 1 - \left(\frac{1 - T_R}{1 - T_{br}} \right)^n \quad (14)$$

and λ_b is calculated using the group contribution method Equation (15):

$$\lambda_b = \sum \Delta\lambda_b + \sum \Delta\lambda_{corr} \quad (15)$$

$\Delta\lambda_b$ is the group contribution value of the different groups and $\Delta\lambda_{corr}$ is a correction factor which may be required for some compounds. "a" and "n" are constants. Excepted for alcohols and phenols (where $a = 0.856$ and $n = 1.23$) the values for these constants are respectively 0.160 and 0.20 for most compounds [6], T_{br} is the ratio of the boiling temperature and the critical temperature.

Sato-Riedel method is based on the equation of Sato-Riedel Equation (16) as follow:

$$\lambda = \frac{1.1053 * (3 + 20(1 - T_R)^{2/3})}{(3 + 20(1 - T_{br})^{2/3}) * M_w^{1/2}} \quad (16)$$

The upper temperature limit for Sato-Riedel method is the critical point and thermal conductivity will not be calculated at temperatures above this.

Heat capacity at constant pressure (C_p) estimation

Two methods were selected to estimate the heat capacity at constant pressure. The most accurate method was chose by comparing the values of the estimated properties with experimental values of this study. Zong *et al.* [43] developed an approach based on chemical constituent fragments to estimate the thermo physical properties. The heat capacity C_p^l can be estimated by Equation (17) ac-

ording to this approach.

$$C_P^l = \sum_A N_{frag,A} C_{P,A}^l(T) \quad (17)$$

where $C_{P,A}^l = A_{1,A} + A_{2,A}T$ (18), $A_{1,A}$ and $A_{2,A}$ are parameters of temperature dependent correlation for fragment A and T is the temperature (K), and $N_{frag,A}$ is the number of fragments A in the component. The detail of Zong *et al.* method and the others parameters can be found in the following references [38] [43].

Ceriani *et al.* [44] applied group contribution method to predict heat capacity for fatty compounds and oils. The equation used is given by Equation (18)

$$C_{Pi}^l = \sum_k N_k \cdot (A_k + B_k T) \quad (18)$$

where N_k is the number of group k in the molecule and A_k , B_k are parameters obtained from the regression. The detail of Ceriani *et al.* [38] [44] method and the others parameters can be found in the following literature [39].

No temperature limit was found for Ceriani *et al.* and Zong *et al.* methods.

3. Results and Discussion

For all the physical properties, the Average Relative Deviation (ARD) which formula given by Equation (19) is used to evaluate the accuracy of the different studied methods and for the validation of the estimated values.

$$ARD(\%) = \frac{1}{N} \sum_1^N 100 * |Exp_v - Est_v| / Exp_v \quad (19)$$

where N is the number of data points, Exp_v is experimental value, Est_v is estimated value.

3.1. Results for Critical Properties and Normal Boiling Point

Table 5 shows the results of critical properties and normal boiling point predicted by the MG method for rapeseed and Jatropha oils.

To test the reliability of MG method, the estimated values of this study have been compared with literature data. However, there are literature data for only canola oil which is another variety of rapeseed oil and then is used for comparison.

Table 5. Rapeseed and Jatropha oil estimated critical properties and normal boiling point by MG method.

| Physical Properties | | T_b (K) | T_c (K) | P_c (bar) | V_c (cm ³ /mol) |
|---------------------|----------------------|-----------|-----------|-------------|------------------------------|
| | This study* | 638.01 | 811.02 | 14.17 | 1.05 |
| Rapeseed oil | Literature [36] [45] | 626.10 | 818.95 | 12.85 | 1.04 |
| | ARD (%) | 1.90 | 0.96 | 10.27 | 1.25 |
| | This study* | 634.51 | 838.39 | 14.35 | 1.03 |
| Jatropha oil | Literature [36] | 623.60 | 837.47 | 13.02 | 1.03 |
| | ARD (%) | 1.75 | 0.10 | 10.21 | 0.68 |

*values of parameters calculated by the authors using the Marrero-Gani's method.

As shown in **Table 5**, the relative deviation between literature data and estimated values for rapeseed and jatropha oils are low for T_b , T_c and V_c confirming the reliability of MG method. Large deviations were only observed for critical pressure in both cases. Poling *et al.* [39] have already noted that for the estimation of the critical pressure, the largest errors are found for the heavier molecules, consequently the estimations may be too high or too low with no obvious pattern for errors under these conditions. This could therefore explain the high ARD for vegetable oils, since they are formed of triglycerides which are large molecules.

3.2. Experimental Results of Rapeseed and Jatropha Oils Physical Properties

The experimental data of this work for physical properties of rapeseed and Jatropha oils, for the temperature range 298 to 523 K, using the different methods and devices above mentioned are given in **Table 6**.

Figure 1 and **Figure 2** show curves evolution of experimental density, heat capacity, thermal conductivity and dynamic viscosity. Figures indicate that all physical properties of the two oils above mentioned decrease as the temperature increases except the heat capacity which increases along with temperature. For the viscosity, the effect of temperature is related to the decrease of intermolecular forces, making easier the flow and therefore the reduction of viscosity [46]. In

Table 6. Experimental values for secondary physical properties of rapeseed and jatropha oils obtained in this work.

| Physical property | Density (kg/m ³) | | Dynamic viscosity (mPa-s) | | Thermal conductivity (W/m.K) | | Heat capacity (J/g.K) | |
|--------------------|------------------------------|--------------|---------------------------|--------------|------------------------------|--------------|-----------------------|--------------|
| Mesasurement error | 0.051 | | 0.10 | | 0.012 | | 0.037 | |
| Temperature (K) | rapeseed oil | Jatropha oil | rapeseed oil | Jatropha oil | rapeseed oil | Jatropha oil | rapeseed oil | Jatropha oil |
| 298 | 911.462 | 914.99 | 134.868 | 88.839 | 0.1654 | 0.1681 | 2.0294 | 2.0373 |
| 303 | 908.117 | 911.294 | 97.7675 | 63.5536 | 0.1648 | 0.1670 | 2.0313 | 2.0642 |
| 323 | 894.735 | 896.51 | 39.6956 | 24.8646 | 0.1617 | 0.1630 | 2.0739 | 2.1903 |
| 343 | 881.353 | 881.726 | 21.9229 | 13.4007 | 0.1587 | 0.1591 | 2.1512 | 2.3228 |
| 363 | 867.971 | 866.942 | 14.0706 | 8.4454 | 0.1559 | 0.1555 | 2.2398 | 2.4377 |
| 383 | 854.589 | 852.158 | 9.8750 | 5.8414 | 0.1533 | 0.1521 | 2.3227 | 2.5197 |
| 403 | 841.207 | 837.374 | 7.3539 | 4.2976 | 0.1508 | 0.1489 | 2.3891 | 2.5622 |
| 423 | 827.825 | 822.59 | 5.7129 | 3.3041 | 0.1485 | 0.1460 | 2.4345 | 2.5674 |
| 443 | 814.443 | 807.806 | 4.5808 | 2.6254 | 0.1464 | 0.1433 | 2.4605 | 2.5458 |
| 463 | 801.061 | 793.022 | 3.7645 | 2.1402 | 0.1444 | 0.1408 | 2.4750 | 2.5171 |
| 483 | 787.679 | 778.238 | 3.1551 | 1.7807 | 0.1426 | 0.1385 | 2.4920 | 2.5094 |
| 503 | 774.297 | 763.454 | 2.6872 | 1.5067 | 0.1409 | 0.1364 | 2.5319 | 2.5595 |
| 523 | 760.915 | 748.67 | 2.3195 | 1.2927 | 0.1394 | 0.1346 | 2.6212 | 2.7129 |

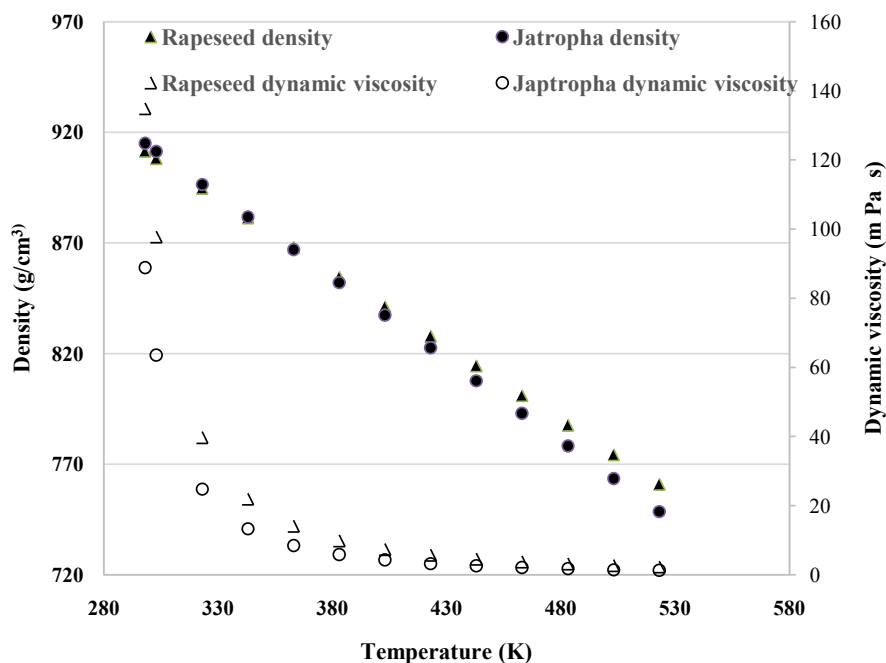


Figure 1. Experimental density and dynamic viscosity curves evolution of rapeseed and Jatropha versus temperature.

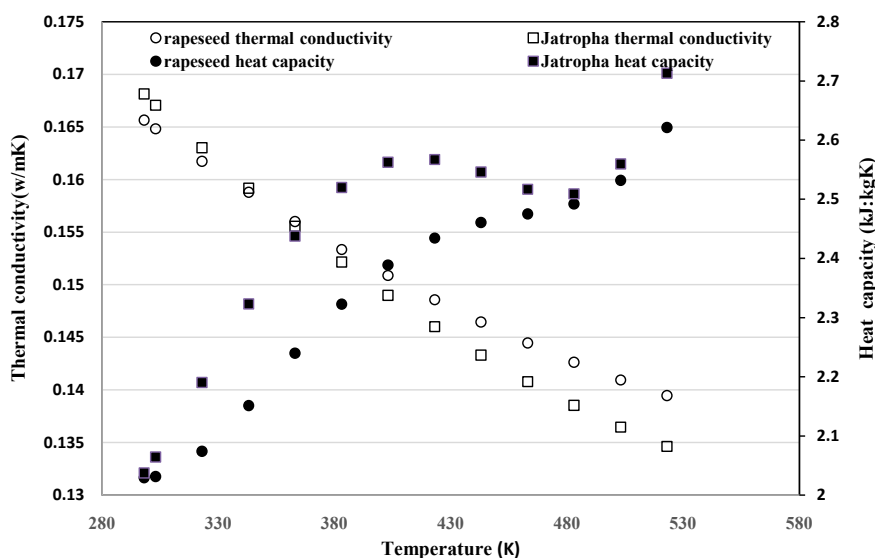


Figure 2. Experimental thermal conductivity and heat capacity curves evolution of rapeseed and Jatropha versus temperature.

the case of density, when the temperature increases, the molecules disperse and the fluid expands in occupying a larger space. As the mass of the fluid remains identical, this expansion causes a decrease in the density. However, the specific heat of the two oils increases along with increasing temperature. This trend confirms the experimental results obtained by Morad *et al.* [40] with DSC method. They also found that this increase in heat capacity with temperature is related to the mobility of the molecules depending on the temperature.

3.3. Estimated Physical Properties for Rapeseed and Jatropha Oils

3.3.1. Density

Figure 3 and Figure 4 show respectively the estimated and experimental density values for rapeseed and Jatropha oils as a function of temperature. In both cases, the estimated and experimental data show the same trend with temperature: density decreases when temperature increases. In particular when temperature increases the Gunn Yamada density curve gets closer to the experimental one while the Ihmels *et al.*'s density curve is closer to the experimental one at low temperatures and deviates more and more as the temperature increases. In addition, low deviations between experimental values and predicted one were found with the two methods as shown in Table 7 showing the goodness

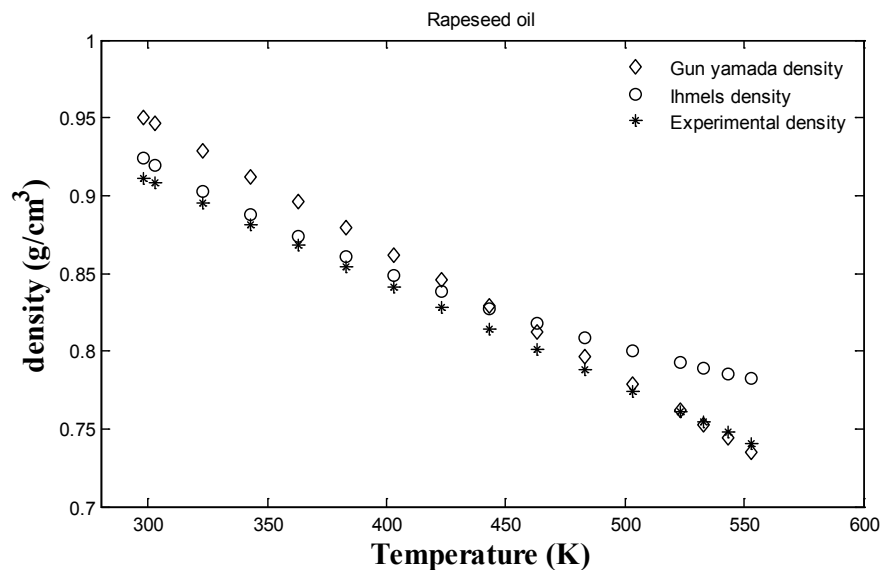


Figure 3. Experimental and estimated density of rapeseed oil.

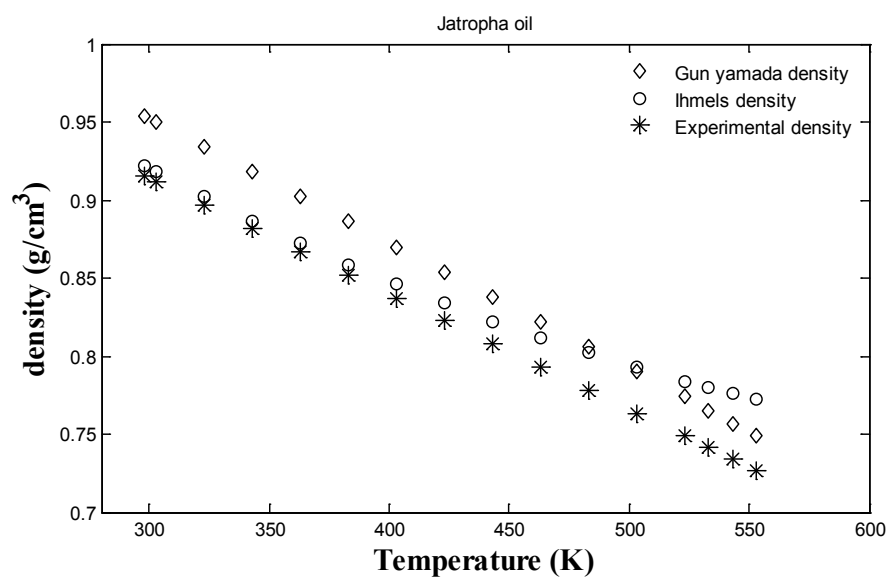


Figure 4. Experimental and estimated density of jatropha oil.

Table 7. ARD values for the different methods used.

| Property | Estimation method | ARD (%) | | data points |
|----------------------|-------------------------------|--------------|--------------|-------------|
| | | Rapeseed oil | Jatropha oil | |
| Density | Gunn Yamada | 2.05 | 3.73 | 16 |
| | Ihmels <i>et al.</i> Gmehling | 2.30 | 2.45 | 16 |
| Dynamic Viscosity | Jöback-Lydersen | 28.39 | 20.42 | 14 |
| | Morris | 32.42 | 58.93 | 14 |
| Thermal conductivity | Sastri | 5.05 | 1.29 | 14 |
| | Sato-Riedel | 34.30 | 39.32 | 14 |
| Heat Capacity | Zong <i>et al.</i> | 6.18 | 7.75 | 14 |
| | Ceriani <i>et al.</i> | 19.03 | 12.18 | 14 |

of the two correlatives methods. However, in view of the purpose of this work, Gunn Yamada correlative method is recommended for extrapolation of density at high temperatures typically observed during the injection phase in diesel engine. Ihmels *et al.*'s method is then recommended for determined edible vegetable oils properties for food purposes.

3.3.2. Dynamic Viscosity

The accuracy of the experimental measurements by considering the imperfections of the geometry and the precision of the rheometer was estimated to be in the order of 10%. Two estimation methods have been discussed. In **Figure 5** and **Figure 6**, calculated dynamic viscosities are compared with experimental values for rapeseed and jatropha oils.

For temperatures lower than 350 K, large errors result, as illustrated on both figures for the two methods. This is due to the fact that Jöback-Lydersen and Morris viscosity correlations [39] [41] don't assume that \ln is a linear function of reciprocal temperature. Because generally for a temperature range from the freezing point to the normal boiling temperature, when the natural logarithm of dynamic viscosity is assume to be a linear reciprocal absolute temperature, good approximation is found.

Therefore, for low temperatures, deviations can be observed. In the same range of temperature, experimental values of viscosity are higher than estimated ones obtained by Jöback-Lydersen and Morris. The two methods give similar accuracies and tend to underestimate vegetable oil dynamic viscosity but the method of Jöback-Lydersen yields the smallest errors.

When the temperature increases from 350 K, relative deviations become smaller and the experimental and estimated curves tend to overlap, especially with the method of Jöback-Lydersen. For higher accuracy, the Jöback-Lydersen method can be selected. In the literature [39] there are generally distinct methods of correlations or viscosities at low temperatures and for high temperature. However, the problem lies in the impossibility of combining the two estimated viscosities.

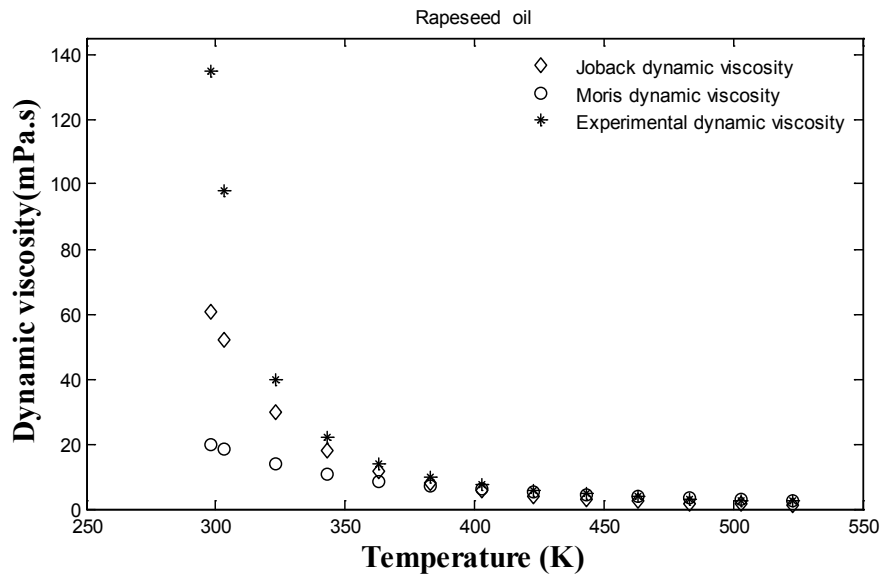


Figure 5. Experimental and estimated dynamic viscosity of rapeseed oil.

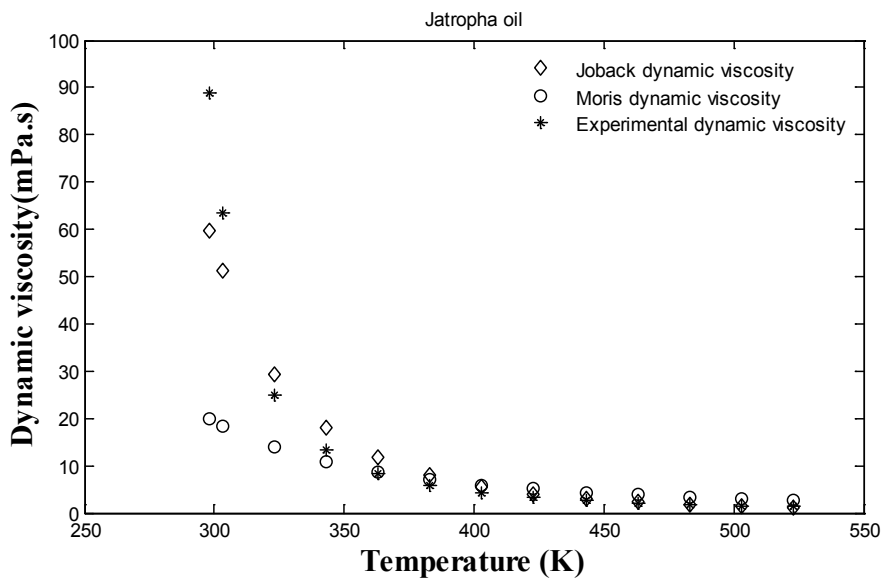


Figure 6. Experimental and estimated dynamic viscosity of jatropha oil.

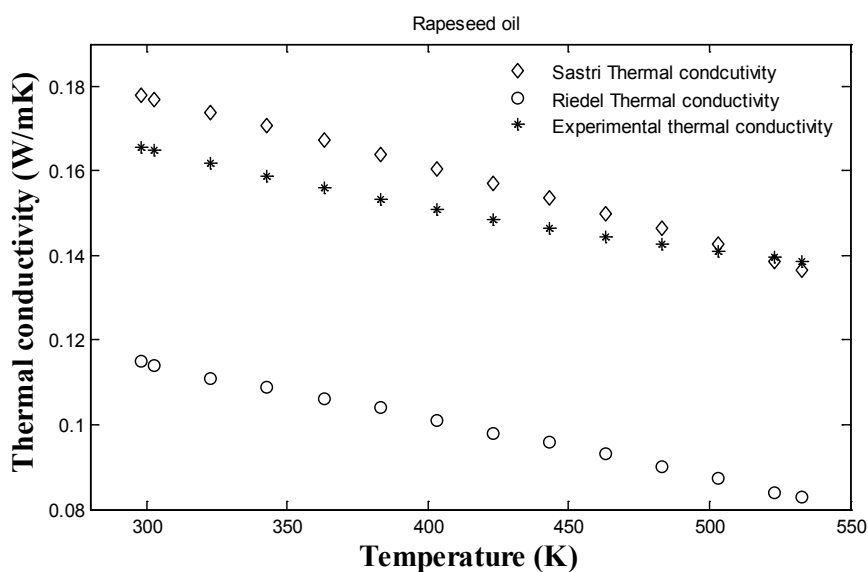
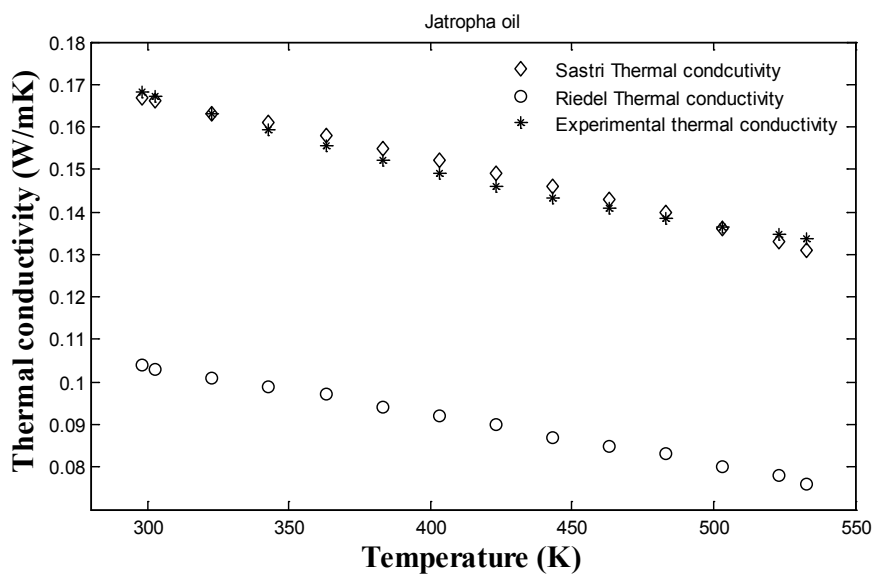
3.3.3. Thermal Conductivity

Table 8 shows the reproducibility error of the device for measuring the thermal conductivity of vegetable oils. The measuring device was found to lead to a reproducibility error of about 1.2%.

Two estimation methods have been discussed: Sastri and Riedel methods [39]. In Figure 7 and Figure 8, calculated thermal conductivity is compared with experimental values for rapeseed and jatropha oils. As expected, in both figures, the results show that the thermal conductivity decreases when the temperature increases. However, the difference between the thermal conductivity obtained by the two estimative methods is very large. The average relative deviation between the estimated thermal conductivity and the experimental values is about 5% for

Table 8. Reproducibility errors for measuring the thermal conductivity of vegetable oils.

| Method | Error (%) | Oil | Reference |
|------------------|-----------|--------------------|-----------|
| Photoacoustic | 2 | Sunflower | [47] |
| | 3 | Soybean | |
| | 1 | Sunflower (21°C) | |
| Thermal analyser | 1,8 | Sunflower (68.7°C) | [48] |
| | 1.2 | Soybean (21°C) | |

**Figure 7.** Experimental and estimated thermal conductivity of rapeseed oil.**Figure 8.** Experimental and estimated thermal conductivity of jatropha oil.

Sastri method. Riedel method overestimates the rapeseed and jatropha oils thermal conductivity. Then, Sastri method gives good agreement between pre-

dicted thermal conductivity and experimental thermal conductivity data in the studied temperature range. The higher agreement of Sastri method with experimental values is probably due to the fact that this correlative method involves the contribution of functional groups as well as correction factors, whereas the Riedel method involves only a reference value and a reduced temperature.

3.3.4. Heat Capacity at Constant Pressure (C_p)

Two methods for estimating liquid heat capacities were considered. **Figure 9** and **Figure 10** show comparison of estimated values and experimental data of heat capacity for rapeseed and jatropha oils. For all the considered cases, the heat capacity increases along with the temperature. The figures show that Zong *et al.*

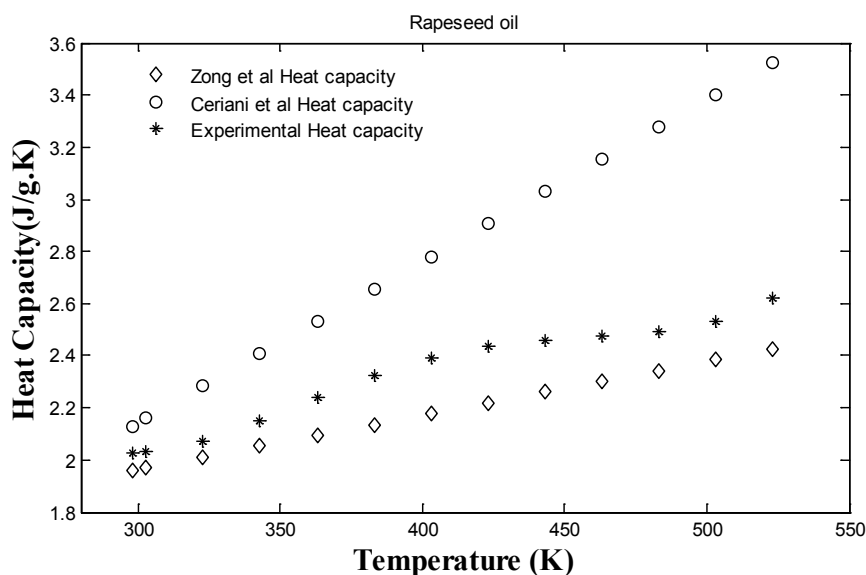


Figure 9. Experimental and estimated thermal heat capacity of rapeseed oil.

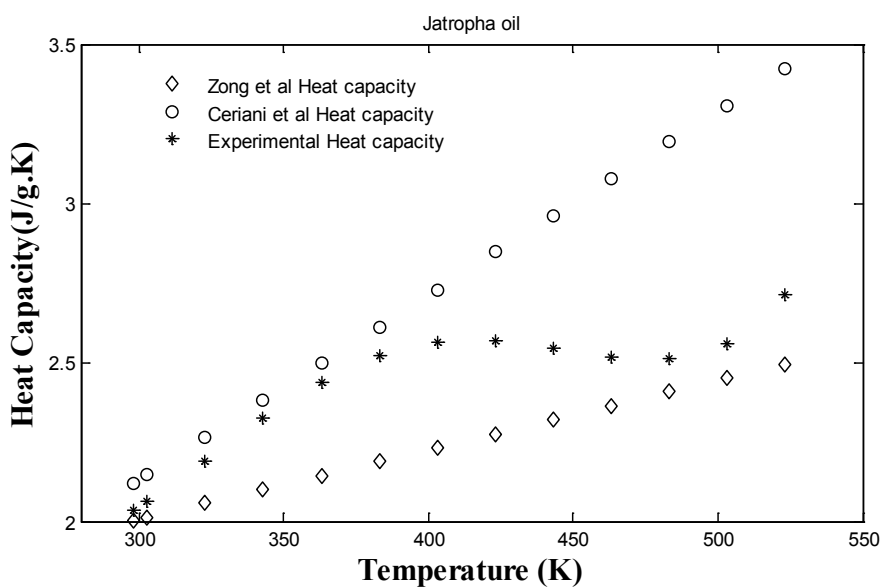


Figure 10. Experimental and estimated thermal heat capacity of jatropha oil.

[43] method give large negative deviation, while Ceriani *et al.* method has small positive deviation [44]. Ceriani's method gives very low errors especially for relatively low temperatures (<400 K). But in the considered temperature range, Zong *et al.* method shows satisfactory agreement with experimental data for both vegetable oils. This can be seen in **Table 7** for ARD values. Zong's method is based on a fragmentation approach of the molecule which is likely to occur at higher temperature whereas Cerani's method is based on the functional groups. This shows that for low temperatures, the functional group approach is better than that of the fragmentation approach. But both of these methods yield better results which show that although the vegetable oils are formed of triglycerides which are themselves formed of fatty acids, their physical properties can be determined successfully by fragmentation or group contribution approach.

In addition, while the curve of the three methods evolves linearly, the experimental one varies first linearly and then changes the slope from 450 K. This clearly shows that from a certain temperature the nature of the oil evolves. This confirms the results of our previous studies which showed that, starting from a certain temperature depending on the oil, the nature of the latter evolved following the thermal decomposition of the triglycerides it contains.

For each recommended estimation model, the curves of the physical properties as well as the ARDs of the two oils can differ depending on the physical property under consideration. The rapeseed and jatropha oils are substantially different in their fatty acid composition. Indeed, according to **Table 2**, although both oils are mono-unsaturated, rapeseed oil contains more than 8% of linolenic acid while jatropha oil does not contain any. This difference in terms of fatty acid composition certainly affects the physical properties of vegetable oils as well as their evolution with temperature. This is in accordance with literature results [49].

4. Conclusions

This work gives tools to overcome the difficulty to determine experimentally physical properties for vegetable oils within the range of temperature typically observed during the injection phase in diesel engine. Based on experimental physical properties of pure vegetable oils determined in this study and existing theoretical models, this work has shown that, within the range 298 to 523 K, rapeseed and jatropha oils' physical properties can be satisfactorily predicted as a function of temperature using group-contribution approach. In this temperature range, it was found that for the prediction of oil density, the Gunn Yamada method was the most accurate, and in line with our experimental data, with an ARD of 1.34 for Rapeseed oil and 0.04 for Jatropha oil. Dynamic viscosity was found to be well-predicted by the Jöback-Lyderson method above 350 K. The calculated ARD of 28.39 for Rapeseed oil and 20.42 for Jatropha oil is much higher because of the large deviation observed at lower temperatures. Thermal conductivity and Heat Capacity were respectively found to be well predicted by

Sastri and Zong *et al.* methods with ARD lower than 7.75 for both Rapeseed and Jatropha oils.

Further studies to be conducted on vegetable oils having extreme fatty acid composition will allow correlating more specifically the evolution up to 523 K of a given physical property to the composition of oils.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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Nomenclature

| | |
|---|--|
| ARD : Average relative deviation, % | T_{ref} : Reference temperature, K |
| C_{lp} : Liquid phase specific heat | V_c : Critic volume, cm ³ /mol |
| C_p : Heat capacity | W : Acentric factor |
| C_p° : Ideal gas heat capacity, kj/kmol K | x_i : Mole fraction of component i |
| E_v : Estimated value | Z_{RAi} : Rackett parameter of component i |
| F_c : Correction factor | $\Delta\lambda_b$: Thermal conductivity group contribution |
| L_v : Litterature value | $\Delta\lambda_{cor}$: Thermal conductivity correction factor |
| M_W : Molecular weight | $\Delta\mu_a$: Jöback groups' contributions a |
| M_{Wi} : Molecular weight of component i | $\Delta\mu_b$: Jöback groups' contributions b |
| N : Number of data points | $\Delta\mu_M$: Group contributions factors |
| T_{br} : Quotient of the boiling temperature, K | λ_0 : Reference thermal conductivity |
| N_{frag} : Fragment number | λ_L : Liquid thermal conductivity |
| N_k : Number of group k | μ^* : Compound class group contribution |
| P_{ci} : Critical pression of component i | μ_L : Liquid dynamic viscosity |
| R : Universal gas constant | ρ_{oil} : Density of vegetable oil |
| T_R : Reduced temperature, K | ρ_{ref} : Density at reference temperature |
