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Density, viscosity, and surface tension of five vegetable oils at elevated temperatures: Measurement and modeling

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ABSTRACT

Density, surface tension and viscosity of five food oils were experimentally measured using the Archimedean method, Pendant drop method, and Brookfield viscometer respectively. Measurements were performed from 23 ± 1°C to the oils' smoke point at intervals of every 20°C. Density and surface tension decreased linearly with increasing temperature, whereas the viscosity decreased exponentially. Density was modeled using the modified Rackett equation, surface tension using the Eötvös equation, and viscosity by the modified Andrade equation. The oil type influenced the density and viscosity of oil, but did not affect surface tension.

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Vegetable oils; High temperature; Density; Viscosity; Surface tension; Modeling

Introduction

Deep fat frying is one of the oldest and the most commonly used techniques to process foods, which involves submerging a food in hot oil (150–200°C) for a short period of time until it reaches a safe minimum internal temperature. [1] It is a relatively low cost process in which the combination of temperature and heat affect the organoleptic properties of food such as flavor, texture, and color. [2,3] Frying is a simultaneous heat and mass transfer process which causes dehydration of the product due to high oil temperature, thus, resulting in a product which has a porous and crispy exterior and a soft and moist interior. [4] The main reactions involved in this process are starch gelatinization, protein denaturation, aromatizing, and coloring via Maillard reactions, rapid cooking, and texture and flavor development. [5–7] Frying also causes changes in the flavor and stability of oils by hydrolysis, oxidation, and polymerization. [6,8] The most commonly used oils for frying include: Canola, corn, cottonseed, olive, palm, peanut, soybean, safflower, and sunflower. [9] Selection of oil for frying is based on long frying stability, fluidity, bland flavor, low tendency to foam or form smoke, low tendency to polymerize, oxidative stability of the oil in the fried food during storage, and the cost. [9–11]

According to the US Department of Agriculture [12], world vegetable oil consumption has increased from 151.68 to 177.16 million metric tons from 2011/12 to 2015/16. Out of the total production, 73.4% of the oil is used for food purposes [12]; 20% of which is for shallow and deep fat frying. [9] Fat plays an important role in enhancing the flavor of foods. Frying of food gives it a unique-flavor and texture combination which is very desirable to the consumers, making fried foods one of the most popular products. [11] However, oil consumption poses significant health problems such as coronary heart diseases, cancer, diabetes, and hypertension, and is irreconcilable with consumer's awareness toward the consumption of healthier and low fat food products. [13] There is an increasing consumer trend for less greasy and healthier products. [7] Considering the health

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impact and the popularity of fried foods, it is important to gain a better understanding of the principles and factors governing oil absorption so that they can be better controlled.

Deep fat frying is one of the most complex transport problems in the food industry. The main mechanisms proposed to govern the oil absorption during frying are water escape and oil uptake; capillary pressure; vapor pressure and vacuum effect; diffusion, adherence and drainage of oil on the food surface. [7,14] Heat is transferred by convection from the oil to the product surface followed by conduction from the surface to the interior of the product. [15] Mass transfer during frying is characterized by the simultaneous movement of water in the form of vapor from the food into the oil, and the movement of oil into the food. [1] Several factors such as moisture content, crust microstructure, product geometry, frying temperature and time, oil type, oil aging and pre and post frying treatments have shown to affect oil absorption during frying. [7,11,16] Frying is known to affect the physical properties of oil such as density, viscosity and surface tension which can in turn affect the transport rate of oil, and thus the rate of oil absorption. Hence, the focus of this study is to understand the change in the oil physical properties at high temperatures, to better predict the effect of parameters affecting oil absorption at frying temperatures.

Density is an important factor which influences oil absorption as it affects the drainage rate after frying and also the mass transfer rate during the cooling stage of frying. [17,18] Density has been experimentally shown to be linearly dependent on temperature. [19,20] However, there is no mathematical equation to predict the effect of temperature on density, especially at high temperatures at which frying is conducted. Frying occurs at temperatures around 180°C; however the densities of commonly used oils are known till around 110–140°C. [19,20] Hence to accurately determine transport rates, density need to be determined and modeled at the frying temperatures. A liquid pycnometer [19], hydrometer with temperature controller [20] are the most commonly used methods to determine density of a liquid at temperatures above room temperature. Both the methods involve use of glass apparatus, hence glass expansion coefficient needs to be accounted for. Also, the assembly of the apparatus is complex. Archimedean method has been successfully used to determine density of liquids at high temperatures (molten igneous rocks) [21]; however it has not been used to measure densities of food oils.

Ziaiifar et al. [7] reported that the viscosity of oil is one of the main factor which governs oil absorption and drainage. The higher is the oil viscosity, the slower is the oil drainage. Oil viscosity depends on oil type as well as frying temperature and oil quality. Kim et al. [13], Yilmaz [18], Esteban et al. [20], and Noureddini et al. [22] measured viscosities of vegetable oils from room temperature up to maximum of 130°C using a glass capillary viscometer. Correlations were developed for viscosity with temperatures using empirical equations as well as relating fatty acid composition to the viscosity change. These studies give valuable information on the effect of temperature on oil degradation but fail to develop models which can be used as predictors of oil viscosity, especially at the frying temperatures. Viscometer and Rheometer are the most commonly used equipment's to measure oil viscosity at high temperatures. [7,18,20,23] Modeling viscosity at frying temperatures can give a better understanding of the flow behavior of the oil during drainage, and thus help to characterize oil absorption.

Surface and interfacial tension (IFT) play a key role in mechanisms of oil uptake, as frying oil moves into the food's tiny pores, which are filled with water and/or steam. [4,24] Surface tension of oils is measured at room temperature, however, frying process is conducted at temperatures between 150–180°C. For many pure liquids, an increase in temperature causes a linear decrease in the surface tension. Investigation of surface and interfacial tensions at high temperature is thus a key element required to characterize and predict the oil absorption by the capillary mechanism. Kalogianni et al. [23] measured the interfacial tension of olive oil and palm oil at room temperature after repeated frying cycles using the pendant drop technique, and found a dynamic decrease in interfacial tension values for both oil types. Thus, frying has a significant effect on surface tension of oil and the effect needs to be predicted at high temperatures to gain a better understanding of oil absorption. Surface tension can be measured by many methods including: Pendant drop, Du Nouy ring, Wilhelmy plate, Maximum bubble pressure, and Contact angle. [25] The Pendant drop technique is commonly used

to measure the IFT of hydrocarbons and crude oils at high temperature [26,27] but has not been used to measure the surface tension of food oils at high temperatures.

While oil viscosity and density as affected by temperature and oil composition have received some attention, the surface tension of oils at frying temperatures in pure or mixed systems has not been studied to a great extent. [28] The current study seeks to address this gap. The objectives of this study are to determine and mathematically model surface tension, density and viscosity of commonly used food oils at high temperatures. The models developed will help to gain a better understanding of oil absorption at the frying temperatures.

Materials and methods

Materials

Canola oil (Wesson, ConAgra Foods Inc.; Omaha NE, USA), extra virgin olive oil (Filippo Berio, SALOV North America Corp.; Lyndhurst NJ, USA), soybean oil (Crisco, The J.M. Smucker Co.; Orrville OH, USA), peanut oil (LouAna, Ventura Foods, LLC; Brea CA, USA) and corn oil (Mazola, ACH Food Companies, Inc.; Memphis TN, USA) were purchased from a local market. Decane was purchased from TCI America (Portland OR, USA). Acetone was purchased from VWR (Radnor, PA, USA). Stainless steel wire (diameter: 0.76 mm) was bought from Hobart Welders (Northern Tool; Raleigh NC, USA). A stainless steel ball (diameter: 25.4 mm) was obtained from the Precision Instrument Machine Shop (North Carolina State University Raleigh, NC, USA).

Density measurements

Oil density was determined by the Archimedean method as described by White. [21] This method uses a solid object of known volume and mass suspended in the test liquid by a wire hanging from a scale. A stainless steel ball welded to a stainless steel wire was used as the reference material for this study. The density (ρ) was calculated using the following equation:

$$\rho = \frac{\beta + s}{V^0 + v'} \quad \text{(Eq. 1)}$$

where β is the buoyancy, V^0 is the volume of the reference object, s is the surface tension effect between the liquid and the wire, and v' is the volume of immersed wire. For the purpose of this study, s was assumed negligible, V^0 was determined to be 8.58 cm^3, and v' equal to 0.01 cm^3. The buoyancy was determined by:

$$\beta = m_v - m_f \quad \text{(Eq. 2)}$$

where m_v and m_f are the mass of the reference object plus immersed wired suspended *in vacuo*, and in the test fluid respectively. The value determined for m_v was 69.67 g. Measurements were performed at room temperature (22 ± 1°C), 40°C and at increasing intervals of 20°C until the smoke point of each oil was reached (200°C; except for olive oil, 180°C). [29,30] Prior to each measurement, the oil was placed inside a beaker and heated under constant agitation until the desired temperature was reached. To maintain a uniform temperature, the wire/ball was also heated along with the oil in the beaker. Measurements were performed in triplicate.

Surface tension measurements

The surface tension of the five vegetable oils in air was determined by pendant drop method using a KRÜSS (Model- DSA30B, KRÜSS GmBH, Hamburg, Germany) as well as ramé-hart goniometer (ramé-hart, Advanced Goniometer, model 300, Succasunna, NJ). The surface tension was measured using Drop Shape Analysis software and DropImage software respectively. An elevated temperature

syringe with a 22 gauge stainless steel needle was used to form the drops inside an environmental chamber equipped with a temperature control system (P/N 100–11, P/N 100–10-20, P/N 100–10-12–22, and P/N 100–07 respectively; ramé-hart instrument co.; Succasunna NJ, USA) with air as the surrounding medium. The environmental chamber and the high temperature syringe, both equipped with a temperature control system (P/N 100–50; ramé-hart instrument co.; Succasunna NJ, USA) and SOLO Temperature Controller Configuration software (AutomationDirect; Cumming GA, USA) were used. Additionally, temperature was monitored at the vicinity of the needle where the drop was generated, using a thermocouple (P/N KMQSS-062U, OMEGA Engineering, Inc; Stamford CT, USA) attached to a data logger with Logger Lite software (LabQuest Mini, Vernier Software & Technology, LLC.; Beaverton OR, USA). Acetone was used to clean all surfaces before and after each experiment to avoid contamination; surfaces were allowed to dry for 1 min before starting a new measurement. Similar to density experiments, measurements were performed at room temperature (22 ± 1°C), 40°C and at increasing intervals of 20°C until the smoke point of each oil was reached (200°C; except for olive oil, 180°C). Three measurements were performed at each temperature.

Viscosity measurements

The dynamic viscosity (μ) of the vegetable oils was determined by a Brookfield viscometer equipped with a thermo-container and programmable temperature controller (LV-DVIII, HT-60, HT-110FR respectively; Brookfield Engineering Laboratories, Inc.; Middleboro MA, USA). The thermo-container was equipped with a sample chamber, and a spindle (HT-2 and SC4-18 respectively; Brookfield Engineering Laboratories, Inc.; Middleboro MA, USA). The system was cooled using a cooling plug assembly (HT-26Y, Brookfield Engineering Laboratories, Inc.; Middleboro MA, USA) attached to pressurized air nozzle. The heating profile for the temperature controller was set up as: heat from 23°C until the smoke point of each oil (200°C, except olive oil 180°C) in increments of 20°C and holding at each temperature for 5 min. Dynamic viscosity measured was then converted into kinematic viscosity (υ, Eq. 12) using the density of each oil, for comparison with data from mathematical models. The measurements were performed at different RPM settings to confirm the Newtonian behavior of the five oils studied (data not shown).

Mathematical modeling

Density

Density as a function of temperature was predicted based on the modified Racket equation (Eq. 3) developed by Spencer and Danner [31] which estimates the molar volume of a saturated pure liquid (V_s):

$$V_s = \left(\frac{RT_c}{P_c}\right) Z_{RA}^{\left[1+(T_r)^{2/7}\right]} \quad \text{(Eq. 3)}$$

where V_s is in cm^3/mol, T_c is the critical temperature (K), P_c is the critical pressure (bar), R is the ideal gas constant ($cm^3bar/molK$), Z_{RA} is the Rackett parameter which is unique to each compound, and T_r is the reduced temperature (K). Halvorsen et al. [32] presented a variation of Eq. (3) to estimate the density of vegetable oils (Eq. 4), which considers the mixture of fatty acids and a correction factor to account for triglycerides.

$$\rho = \frac{\left(\sum x_i MW_i\right)}{R\left(\sum \frac{x_i Tc_i}{Pc_i}\right)\left(\sum x_i Z_{RA_i}\right)^{\left[1+(T_r)^{2/7}\right]}} + F_c \quad \text{(Eq. 4)}$$

where x_i is the mole fraction of each component, MW_i the molecular weight (g/mol) of each component, and F_c is the correction factor. Values of MW_i, T_C, P_C, and Z_{RA} corresponding to each fatty acid present in the composition of each oil used in this study are reported in Table 1. The reduced temperature (T_r) was obtained using the temperature (T) at which the estimation was done and a molar average of the critical temperatures as pseudocritical temperature (Eq. 5). [32]

Table 1. Molecular weight, critical properties and Racket parameters of fatty acids.

Fatty acid	MW $_i$ (g/mol)	$T_{C\ i}$ (K)*	$P_{C\ i}$ (bar)*	$Z_{RA\ i}$*
C16:0	256.43	799.89	14.08	0.2267
C16:1	254.43	800.34	14.71	0.2290
C18:0	284.49	819.00	12.25	0.2205
C18:1	282.49	819.41	12.76	0.2230
C18:2	280.49	819.82	13.31	0.2255
C18:3	278.49	820.23	13.89	0.2284
C20:0	312.54	836.65	10.76	0.2149
C20:1	310.54	837.03	11.18	0.2172
C22:0	340.59	853.06	9.52	0.2095
C24:0	368.65	868.38	8.49	0.2040
C26:0	396.70	882.76	7.61	0.1990

*Obtained from Halvorsen et al. [30]

$$T_r = \frac{T}{(\sum x_i T_{C_i})} \quad \text{(Eq. 5)}$$

The correction factor (F_c) ranges from 0.02 to 0.04 and only depends on oil type, more specifically the molecular weight. When the molecular weight of the oil (MW_{oil}) was lower than 875 g/mol, F_c was calculated using Eq. (6); however, when it was higher, Eq. (7) was used. MW_{oil} was calculated based on the fatty acid composition using Eq. (8).[32]

$$F_C = 0.0236 + 0.000082\ |875 - MW_{oil}| \quad \text{(Eq. 6)}$$

$$F_C = 0.0236 + 0.000098\ |875 - MW_{oil}| \quad \text{(Eq. 7)}$$

$$MW_{oil} = 3\sum x_i MW_i + 38.0488 \quad \text{(Eq. 8)}$$

Surface tension

Surface tension was predicted by the Eötvös equation (Eq. 9) as a direct function of temperature for data obtained from both the goniometers:

$$\gamma_{LG}\left(MW_{oil}/\rho_{oil}\right)^{2/3} = K_E\left(T_{C,oil} - T\right) \quad \text{(Eq. 9)}$$

where γ_{LG} is the liquid-gas surface tension (mN/m), MW_{oil} is the oil's molar mass (g/mol), ρ_{oil} is the oil's density (g/cm^3), K_E is the Eötvös constant ($dynes\ cm/mol^{2/3}\ K$), T_C is the oil's critical temperature (K), and T is the temperature (K) at which γ_{LG} is estimated. K_E is the measure of entropy of the surface, and was determined using the method of least squares for each of the systems. It is known that geometrically complex molecules have higher K_E values (e.g., tripalmitine, $K_E = 5.4$), than those almost spherical molecules (e.g., mercury, $K_E \approx 1$). [33] Hence, K_E values were determined for each oil type. The surface tension data was also modeled using the predicted density values obtained from the Rackett equation and fitting them in the Eötvös equation.

Viscosity

Viscosity as a function of temperature was modeled using the modified Andrade equation (Eq. 10) which is derived from the Arrhenius equation, and was used by Esteban et al. [20], Noureddini et al. [22], and Yilmaz [18] to understand the viscosities of vegetable oils and fatty acids.

$$v = e^{(a+b/T+c/T^2)} \quad \text{(Eq. 10)}$$

where v is the oil kinematic viscosity (mm^2/s), T is the temperature (K), a, b and c are correlation constants which are calculated using the method of least squares. Accuracy of the mathematical models for each of the physical properties measured, was determined from the percent error.

Statistical analysis

Measurements were performed in triplicate and data was analyzed using Minitab 17 (Minitab Inc., State College PA, USA). Regression (REG) analysis was performed on measured viscosity, density and surface tension for all five oils. Tukey's test ($\alpha = 0.05$) was used to determine differences among measured physical properties with temperature for each oil.

Results and discussion

Effect of temperature and oil type on physical properties of oils

Density

Density of soybean, canola, corn, peanut and olive oil was measured by the Archimedean method from room temperature to 200°C (except olive oil, 180°C) (Table 2). The density values obtained were similar to those previously reported in literature.[19] Regression analysis showed that there was a linear decrease in density with temperature for all oils (Fig. 1). The intercept and slopes for each oil are reported in Table 3. Statistical analysis using General linear model (GLM) showed significant effect ($p < 0.05$) of temperature and oil type on density.

Surface tension

Surface tension of soybean, canola, corn, peanut and olive oil was measured using a KRÜSS and ramé-hart goniometer from room temperature to 200°C (except olive oil 180°C) (Table 4). Regardless of the oil type or the equipment used, there was a significant decrease ($p < 0.05$) in surface tension as the temperature increased; Fig. 2 shows data obtained using the KRÜSS goniometer. The results obtained agree with data previously reported in literature.[34] Regression analysis showed that there was a linear decrease in surface tension with temperature for all oils. The intercept and slopes for each oil are reported in Table 5. There was a difference in values for the surface tension measured with KRÜSS vs ramé-hart equipment mainly because of the numerical method used by each equipment in calculating surface tension from the Young's equation.

Statistical analysis showed significant effect ($p < 0.05$) of temperature and oil type on the surface tension values. However, the difference in surface tension values for different oil types is within the range of accuracy of each equipment and the effect can be ignored. Thus, temperature was the only significant factor affecting surface tension. This finding agrees with the result from Xu et al. [34] where no effect of oil type on surface tension was observed, and the effect could be attributed to the presence of long-acyl chains in all oils, whose surface tensions are not very different from each other.

Viscosity

Viscosity of soybean, canola, corn, peanut and olive oil was measured using a viscometer from room temperature to 200°C (Table 6). All the oils used in the study showed Newtonian behavior at the range of temperatures studied. Regardless of the oil type, there was a significant decrease ($p < 0.05$) in viscosity as the temperature increased (Fig. 3). In the current study, viscosity was found to have a power law relation of the second degree with temperature (Eq. 11), irrespective of the type of oil used.

$$\mu = aT^b \tag{11}$$

where μ is the dynamic viscosity ($mPa \cdot s$), T is the temperature (°C), a is the coefficient, and b is the power of the equation obtained by regression analysis (Table 7). The viscosity values are similar to those obtained by Noureddini et al.[22], Miller et al. [28] and Ziaiifar et al. [7] It was observed that the viscosity values were a quadratic function of temperature for all the oils. Statistical analysis using showed significant effect ($p < 0.05$) of temperature and oil type on the viscosity values; differences between oils were more noticeable at temperatures below 100°C.

Table 2. Comparison of the average density values between five vegetable oils at different temperatures.

	ρ_{oil} (kg/m^3)				
T (°C)	Canola oil	Corn oil	Olive oil	Peanut oil	Soybean oil
22 ± 1	913.3 ± 0.7^{b,c}	915.3 ± 0.7^{a,b}	908.7 ± 0.7^{d}	912.1 ± 0.7^{c}	915.7 ± 0.7^{a}
40	901.7 ± 0.7^{f}	904.4 ± 0.0^{e}	897.4 ± 0.0^{g}	899.3 ± 0.7^{g}	903.3 ± 0.0^{e,f}
60	890.4 ± 0.0^{i}	892.8 ± 0.0^{h}	885.8 ± 0.0^{j}	886.8 ± 0.7^{j}	892.4 ± 0.7^{h,i}
80	878.0 ± 0.7^{l}	880.7 ± 0.7^{k}	874.2 ± 1.2^{m}	875.2 ± 0.7^{m}	880.0 ± 0.0^{k,l}
100	867.2 ± 1.2^{n}	868.3 ± 0.0^{n}	861.7 ± 0.7^{o}	862.4 ± 0.7^{o}	867.6 ± 0.7^{n}
120	854.0 ± 0.7^{p}	855.9 ± 0.7^{p}	849.7 ± 0.0^{q}	850.4 ± 0.7^{q}	855.9 ± 0.7^{p}
140	841.6 ± 0.0^{s}	843.5 ± 0.7^{r,s}	836.9 ± 0.0^{t}	837.6 ± 0.7^{t}	844.7 ± 0.7^{r}
160	829.1 ± 0.7^{v}	832.2 ± 0.0^{u}	825.3 ± 0.0^{w}	825.9 ± 0.7^{w}	833.0 ± 0.7^{u}
180	817.1 ± 0.0^{y}	819.4 ± 0.0^{x}	813.6 ± 1.2^{z}	814.3 ± 0.7^{z}	820.2 ± 0.7^{x}
200	806.6 ± 0.0^{aa}	807.8 ± 0.0^{aa}	–	801.5 ± 0.7^{ab}	807.4 ± 0.7^{aa}

±: corresponds to the standard deviation, $n = 3$.
[a:] Tukey mean comparison per parameter (95% Confidence)- no significant difference between values sharing same letter.

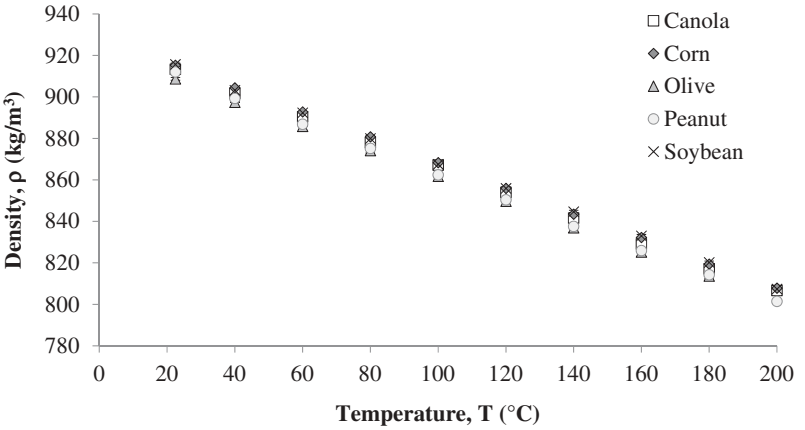


Figure 1. Density values of five vegetable oils from room temperature to the smoke point of each oil.

Table 3. Intercept (ρ_0), slope (ρ_1) and correlation coefficient (r^2) values corresponding to the empirical equation to predict density of each vegetable oil.

Oil type	ρ_0	ρ_1	r^2	Temp range (°C)
Canola	926.7	−0.606	0.999	22–200
Corn	928.9	−0.606	1.000	22–200
Olive	922.2	−0.606	1.000	22–180
Peanut	924.2	−0.615	1.000	22–200
Soybean	928.2	−0.600	0.999	22–200

Mathematical modeling of physical properties of oils at high temperatures

Density

Density was modeled as a function of temperature using the modified Rackett equation (Eq. 4). In order to predict the density of a vegetable oil using this equation, it is necessary to know the critical temperature, critical pressure, molecular weight, and Rackett parameter of each fatty acid present in the oil (Table 1), as well as the molar ratios at which the fatty acids are present. Rackett parameter is estimated using these values and is a measure of the molar volume of a saturated pure liquid. [35] The T_c, P_c and Z_{RA} were calculated as the sum of the corresponding property for each fatty acid present

Table 4. Comparison of the average surface tension values between five vegetable oils at different temperatures measured using KRÜSS and ramé-hart goniometers.

	γ_{oil} (mN/m)				
T (°C)	Canola oil	Corn oil	Olive oil	Peanut oil	Soybean oil
Determined using KRÜSS goniometer					
23 ± 0	31.3 ± 0.3^{b,c}	31.6 ± 0.1^{a,b}	31.9 ± 0.0^{a}	31.3 ± 0.1^{b,c}	31.3 ± 0.4^{b,c,d}
40	30.5 ± 0.2^{e,f}	30.7 ± 0.1^{d,e}	30.9 ± 0.0^{c,d,e}	30.3 ± 0.2^{e,f}	30.6 ± 0.3^{e}
60	29.6 ± 0.2^{g}	29.7 ± 0.1^{g}	30.0 ± 0.0^{f,g}	29.7 ± 0.0^{g}	29.9 ± 0.0^{f,g}
80	28.6 ± 0.2^{h}	28.5 ± 0.1^{h,i}	28.8 ± 0.2^{h}	28.8 ± 0.1^{h}	28.7 ± 0.4^{h}
100	27.7 ± 0.1^{j}	27.7 ± 0.1^{j}	27.7 ± 0.0^{j}	27.7 ± 0.1^{j}	27.9 ± 0.1^{i,j}
120	26.5 ± 0.1^{k}	26.4 ± 0.2^{k,l}	26.6 ± 0.1^{k}	26.5 ± 0.0^{k}	27.0 ± 0.0^{k}
140	25.6 ± 0.2^{m,n}	25.3 ± 0.2^{n}	25.7 ± 0.5^{m,n}	25.5 ± 0.1^{m,n}	25.9 ± 0.1$^{l.m}$
160	24.6 ± 0.1°	24.1 ± 0.1^{o,p}	24.3 ± 0.0^{o,p}	24.5 ± 0.0°	24.5 ± 0.3°
180	23.5 ± 0.0^{q,r}	23.0 ± 0.3^{r,s}	23.1 ± 0.1^{r}	23.0 ± 0.1^{r,s}	23.8 ± 0.1^{p,q}
200	22.4 ± 0.1^{t}	22.0 ± 0.2^{t}	–	21.3 ± 0.4^{u}	22.5 ± 0.1^{s,t}
Determined using ramé-hart goniometer					
23 ± 1	32.4 ± 0.1^{a}	32.2 ± 0.1^{b}	32.0 ± 0.1^{b}	32.2 ± 0.1^{a,b}	29.4 ± 0.0^{g}
40	31.3 ± 0.1^{c}	30.6 ± 0.1^{e}	30.9 ± 0.1^{d}	30.9 ± 0.0^{d}	28.4 ± 0.1^{i}
60	29.7 ± 0.0^{f}	29.3 ± 0.0^{g,h}	29.5 ± 0.1^{f,g}	29.2 ± 0.0^{h}	27.2 ± 0.1^{l}
80	28.2 ± 0.1^{j}	27.8 ± 0.0^{k}	28.1 ± 0.1^{j}	27.6 ± 0.0^{k}	25.9 ± 0.1^{p}
100	26.7 ± 0.1^{m}	26.3 ± 0.0^{n,o}	26.5 ± 0.1^{m,n}	26.2 ± 0.1°	24.2 ± 0.1^{s}
120	25.3 ± 0.1^{q}	25.0 ± 0.1^{r}	25.1 ± 0.0^{q,r}	25.3 ± 0.1^{q}	23.0 ± 0.1^{v}
140	24.0 ± 0.1^{s,t}	23.7 ± 0.0^{u}	23.7 ± 0.1^{u}	23.9 ± 0.0^{t}	21.8 ± 0.1^{z}
160	22.7 ± 0.0^{w,x}	22.2 ± 0.0^{y}	22.5 ± 0.1^{x}	22.7 ± 0.0^{w}	20.9 ± 0.1^{ac}
180	21.2 ± 0.1^{ab}	21.1 ± 0.0^{ab}	21.1 ± 0.1^{ab}	21.5 ± 0.1^{aa}	19.5 ± 0.1^{ae}
200	20.1 ± 0.1^{ad}	20.0 ± 0.1^{ad}	–	20.0 ± 0.0^{ad}	19.0 ± 0.1^{af}

±: corresponds to the standard deviation, n ≥ 3.
$^{a\text{-}t}$: Tukey mean comparison per parameter (95% Confidence)- no significant difference between values sharing same letter. Tukey mean analysis conducted separately for each equipment.

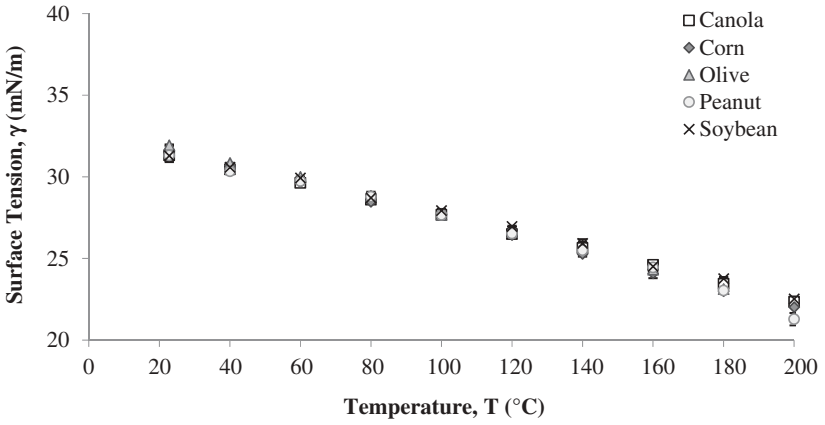


Figure 2. Surface tension values of five vegetable oils from room temperature to each oil's smoke point determined using a KRÜSS goniometer.

in the oil weighted with its correspondent molar fraction; MW_{oil} was calculated using Eq. (9) (Table 8). Similar trends were observed between experimental and predicted data with increasing temperature (Fig. 4). The predicted values and error ($E\%$) at the temperatures at which experiments were conducted for all five oils used in the study are reported in Table 9. The error increased with increasing temperature, but the overall error was not higher than two percent for the temperature range used in the study.

Table 5. Intercept (γ_0), slope (γ_1) and correlation coefficient (r^2) values corresponding to the empirical equation to predict surface tension of each vegetable oil.

Oil type	γ_0	γ_1	r^2	Temp range (°C)
For values determined using KRÜSS goniometer				
Canola	32.58	−0.050	0.996	23–200
Corn	32.92	−0.055	0.997	23–200
Olive	33.20	−0.055	0.994	23–180
Peanut	32.90	−0.055	0.988	23–200
Soybean	32.72	−0.050	0.991	23–200
For values determined using ramé-hart goniometer				
Canola	33.94	−0.071	0.999	23–200
Corn	33.41	−0.069	0.997	23–200
Olive	33.65	−0.070	0.999	23–180
Peanut	33.38	−0.067	0.995	23–200
Soybean	30.74	−0.062	0.993	23–200

Table 6. Comparison of the average viscosity values between five vegetable oils over a range of temperatures.

	μ_{oil} (mPa·m)				
T (°C)	Canola oil	Corn oil	Olive oil	Peanut oil	Soybean oil
22 ± 1	63.5 ± 1.6^{c}	59.2 ± 0.8^{d}	74.1 ± 2.2^{a}	72.2 ± 0.8^{b}	57.1 ± 1.1^{e}
40	34.9 ± 0.9^{h}	34.5 ± 0.6^{h}	40.1 ± 1.5^{f}	38.8 ± 0.7^{g}	31.3 ± 0.6^{i}
60	18.8 ± 0.3^{k}	19.1 ± 0.3^{k}	21.1 ± 0.9^{j}	20.5 ± 0.4^{j}	17.9 ± 0.7^{k}
80	$11.8 \pm 0.4^{m,n}$	$12.5 \pm 0.2^{l,m,n}$	13.4 ± 0.9^{l}	$12.6 \pm 0.4^{l,m}$	11.4 ± 0.6^{n}
100	$8.2 \pm 0.4^{o,p}$	$9.0 \pm 0.2^{o,p}$	9.6 ± 0.8^{o}	$8.8 \pm 0.5^{o,p}$	8.3 ± 0.6^{p}
120	$5.5 \pm 0.2^{q,r,s,t}$	6.4 ± 0.3^{q}	$6.1 \pm 0.2^{q,r}$	$6.1 \pm 0.5^{q,r}$	$5.6 \pm 0.4^{q,r,s}$
140	$4.4 \pm 0.1^{s,t,u,v,w}$	$5.1 \pm 0.2^{r,s,t,u}$	$4.8 \pm 0.2^{s,t,u,v}$	$4.8 \pm 0.5^{s,t,u,v}$	$4.8 \pm 0.7^{s,t,u,v}$
160	$3.7 \pm 0.2^{v,w,x,y,z}$	$4.3 \pm 0.2^{t,u,v,w}$	$4.0 \pm 0.1^{u,v,w,x}$	$4.0 \pm 0.5^{u,v,w,x}$	$3.9 \pm 0.3^{u,v,w,x,y}$
180	$3.0 \pm 0.2^{x,y,z}$	$3.5 \pm 0.1^{w,x,y,z}$	$3.3 \pm 0.1^{w,x,y,z}$	$3.1 \pm 0.2^{w,x,y,z}$	$3.2 \pm 0.3^{w,x,y,z}$
200	2.6 ± 0.3^{z}	$3.1 \pm 0.2^{w,x,y,z}$		$2.7 \pm 0.1^{y,z}$	$2.8 \pm 0.3^{x,y,z}$

±: corresponds to the standard deviation, n ≥ 3.
[a]: Tukey mean comparison per parameter (95% Confidence) no significant difference between values sharing same letter.

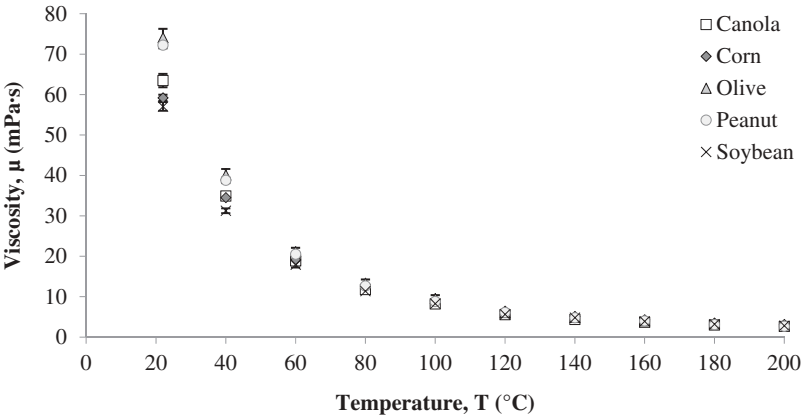


Figure 3. Viscosity values determined for five vegetable oils from room temperature to the smoke point of each oil.

Surface tension

Surface tension was modeled as a function of temperature using the semi-empirical Eötvös equation (Eq. 3). [33,36] It is necessary to know the value of critical temperature of oil and the molecular weight to make predictions using Eötvös equation. Eötvös constant (K_E) is the measure of entropy of the surface and its value was calculated using the method of least squares. The experimental and

Table 7. Parameters *a* and *b*, and correlation coefficient (r^2) values corresponding to the empirical equation to predict viscosity for each vegetable oil.

Oil type	a	b	r^2	Temp range (°C)
Canola	8299.80	−1.52	0.99	22–200
Corn	5188.03	−1.39	0.99	22–200
Olive	10358.12	−1.54	0.99	22–180
Peanut	10463.01	−1.55	0.99	22–200
Soybean	5332.41	−1.42	0.99	22–200

Table 8. Molecular weight, critical temperature, and critical pressure of five vegetable oils.

Oil type	MW_{oil} (g/mol)	$T_{C\ oil}$ (K)	$P_{C\ oil}$ (bar)	$Z_{RA\ oil}$
Canola	881.8 ± 0.0	819.1 ± 0.0	13.0 ± 0.0	0.224 ± 0.000
Corn	873.2 ± 0.0	817.3 ± 0.0	13.2 ± 0.0	0.225 ± 0.000
Olive	874.5 ± 0.5	816.8 ± 0.1	13.0 ± 0.0	0.224 ± 0.000
Peanut	887.5 ± 0.1	819.7 ± 0.0	12.8 ± 0.0	0.223 ± 0.000
Soybean	873.8 ± 0.0	817.7 ± 0.0	13.3 ± 0.0	0.225 ± 0.000

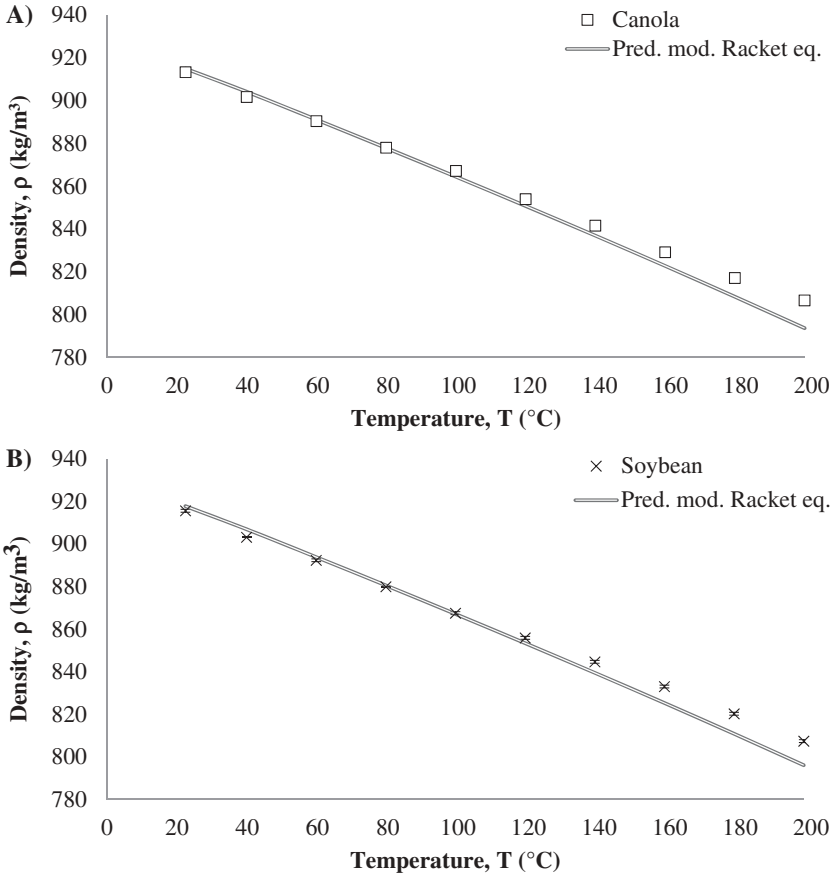


Figure 4. Comparison between density experimental values of A) Canola oil, and B) Soybean oil and their corresponding predicted values by the modified Racket equation.

Table 9. Predicted density values for each vegetable oil by the modified Rackett equation and their corresponding percentage error.

	Canola oil			Corn oil			Olive oil			Peanut oil			Soybean oil		
	ρ_{oil} (kg/m^3)			ρ_{oil} (kg/m^3)			ρ_{oil} (kg/m^3)			ρ_{oil} (kg/m^3)			ρ_{oil} (kg/m^3)		
T (°C)	EXP	PRED	E%	EXP	PRED	E%	EXP	PRED	E%	EXP	PRED	E%	EXP	PRED	E%
22 ± 1	913.3^{b}	915.0^{a}	0.2	915.3^{b}	917.0^{a}	0.2	908.7^{b}	910.9^{a}	0.2	912.1^{a}	910.2^{a}	0.2	915.7^{b}	917.8^{a}	0.2
40	901.7^{d}	904.2^{c}	0.3	904.4^{d}	906.1^{c}	0.2	897.4^{d}	900.1^{c}	0.3	899.3^{b}	899.5^{b}	0.0	903.3^{d}	907.0^{c}	0.4
60	890.4^{e}	891.1^{e}	0.1	892.8^{e}	893.0^{e}	0.0	885.8^{e}	887.0^{e}	0.1	886.8^{c}	886.5^{c}	0.0	892.4^{e}	893.9^{e}	0.2
80	878.0^{f}	877.9^{f}	0.0	880.7^{f}	879.7^{g}	0.1	874.2^{f}	873.8^{f}	0.0	875.2^{d}	873.3^{e}	0.2	880.0^{f}	880.6^{f}	0.1
100	867.2^{g}	864.4^{h}	0.3	868.3^{h}	866.2^{i}	0.2	861.7^{g}	860.3^{h}	0.2	862.4^{f}	859.9^{g}	0.3	867.6^{g}	867.1^{g}	0.0
120	854.0^{i}	850.8^{j}	0.4	855.9^{j}	852.5^{k}	0.4	849.7^{i}	846.6^{j}	0.4	850.4^{h}	846.3^{i}	0.5	855.9^{h}	853.4^{i}	0.3
140	841.6^{k}	836.9^{l}	0.6	843.5^{l}	838.6^{m}	0.6	836.9^{k}	832.7^{l}	0.5	837.6^{j}	832.5^{k}	0.6	844.7^{j}	839.5^{k}	0.6
160	829.1^{m}	822.8^{n}	0.8	832.2^{n}	824.4^{o}	0.9	825.3^{m}	818.6^{n}	0.8	825.9^{l}	818.4^{m}	0.9	833.0^{l}	825.3^{m}	0.9
180	817.1^{o}	808.4^{p}	1.1	819.4^{p}	810.0^{q}	1.2	813.6^{o}	804.2^{p}	1.2	814.3^{n}	804.1^{o}	1.3	820.2^{n}	810.8^{o}	1.1
200	806.6^{q}	793.6^{r}	1.6	807.8^{r}	795.2^{s}	1.6	–	–		801.5^{p}	789.4^{q}	1.5	807.4^{p}	796.1^{q}	1.4

[a:] Tukey mean comparison within each oil type over temperature range (95% Confidence) no significant difference between values sharing same letter.

predicted data were observed to follow a similar trend with increase in temperature (Fig. 5). For all the oils used in the study, K_E of 6.2 dynes·cm/mol$^{2/3}$·K fit the data using the KRÜSS goniometer and K_E of 5.9 dynes·cm/mol$^{2/3}$·K fit the data using the ramé-hart goniometer. Same Eötvös constant values could be used to fit the data for all oils studied with one equipment. The Eötvös constant depends on the geometry of the molecule, and the similarity in values irrespective of oil type was as expected. The difference in Eötvös constant with respect to equipment was mainly because of the differences in numerical method used for surface tension calculation by the equipment, which affected the fit of the model. Models presented in the manuscript are from the data obtained using the KRÜSS goniometer.

The predicted values and error ($E\%$) at the temperatures at which experiments were conducted for all five oils used in the study are reported in Table 10. The error was less than ten percent for all five oils at the temperature range studied; this was true for predicted values compared with data obtained from both equipment, when using their corresponding K_E. According to the data, the Eötvös equation has a higher accuracy of prediction of surface tension between 60–100°C, as the error was less than five percent in this temperature range for all five oils. Overall, the Eötvös equation overestimates the surface tension value at lower temperatures (23 and 40°C) and underestimates the surface tension at higher temperatures (140–200°C). Surface tension was also modeled using a combination of Eötvös and modified Rackett equation (Fig. 5). The model can help to predict

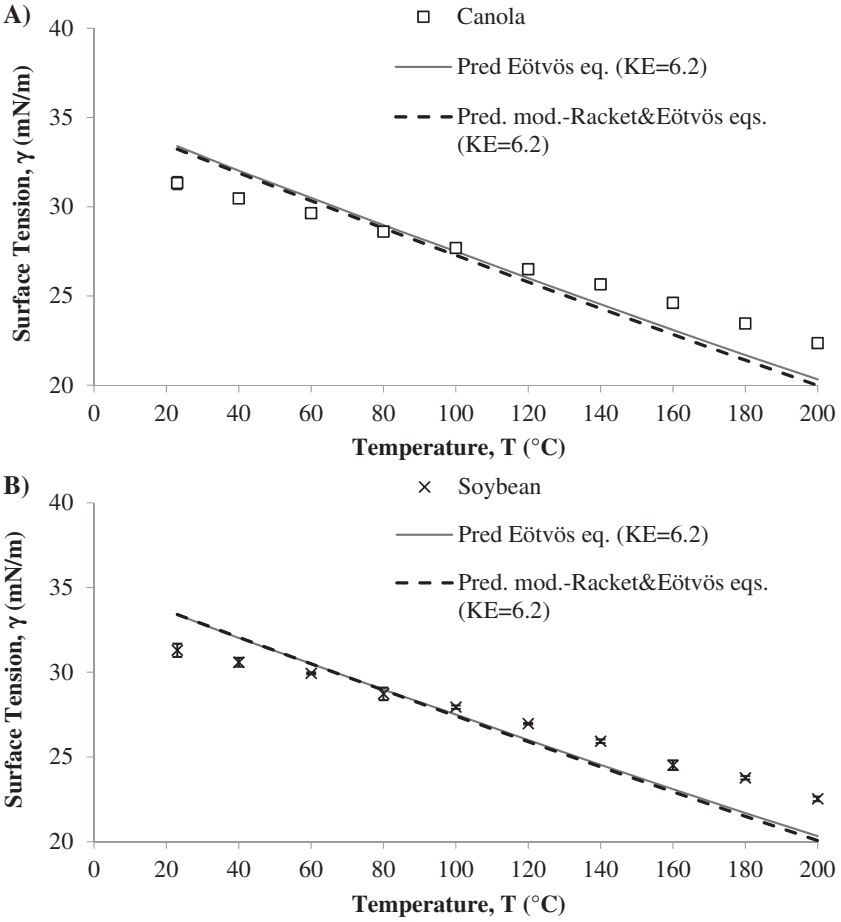


Figure 5. Comparison between surface tension experimental values obtained by KRÜSS goniometer of A) Canola oil, and B) Soybean oil and their corresponding predicted values by the Eötvös equation and the modified Racket- Eötvös equations.

Table 10. Predicted surface tension values for each vegetable oil by the Eötvös equation (K_E = 6.2 dynes·cm/mol$^{2/3}$·K) and their corresponding percentage error comparing with experimental data obtained using KRÜSS goniometer.

	Canola oil			Corn oil			Olive oil			Peanut oil			Soybean oil		
	γ_{oil} (mN/m)			γ_{oil} (mN/m)			γ_{oil} (mN/m)			γ_{oil} (mN/m)			γ_{oil} (mN/m)		
T (°C)	EXP	PRED	E%	EXP	PRED	E%	EXP	PRED	E%	EXP	PRED	E%	EXP	PRED	E%
23 ± 1	31.3^{b}	33.4^{a}	6.6	31.6^{c}	33.6^{a}	6.3	31.9^{b}	33.3^{a}	4.3	31.3^{c}	33.3^{a}	6.2	31.3^{c}	33.6^{a}	7.3
40	30.5^{c}	32.0^{b}	5.1	30.7^{d}	32.2^{b}	4.83	0.9^{c}	32.0^{b}	3.6	30.3^{d}	31.9^{b}	5.0	30.6^{d}	32.2^{b}	5.2
60	29.6^{d}	30.5^{c}	2.9	29.7^{e}	30.6^{d}	3.3	30.0^{d}	30.4^{d}	1.4	29.7^{e}	30.3^{d}	2.0	29.9^{e}	30.6^{d}	2.4
80	28.6^{e}	29.0^{e}	1.3	28.5^{g}	29.1^{f}	2.2	28.8^{e}	28.9^{e}	0.2	28.8^{f}	28.8^{f}	0.1	28.7^{f}	29.1^{f}	1.3
100	27.7^{f}	27.5^{f}	0.7	27.7^{h}	27.6^{h}	0.4	27.7^{f}	27.4^{f}	1.0	27.7^{g}	27.3^{h}	1.3	27.9^{g}	27.6^{g}	1.3
120	26.5^{g}	26.0^{h}	1.9	26.4^{i}	26.1^{j}	1.3	26.6^{g}	25.9^{h}	2.6	26.5^{i}	25.9^{j}	2.6	27.0^{h}	26.1^{i}	3.2
140	25.6^{h}	24.5^{i}	4.3	25.3^{k}	24.6^{l}	2.6	25.7^{h}	24.4^{i}	4.7	25.5^{h}	24.4^{k}	4.4	25.9^{i}	24.7^{j}	4.9
160	24.6^{i}	23.1^{j}	6.1	24.1^{m}	23.2^{n}	3.7	24.3^{i}	23.0^{j}	5.3	24.5^{k}	23.0^{l}	6.0	24.5^{j}	23.2^{l}	5.2
180	23.5^{j}	21.7^{l}	7.5	23.0^{n}	21.8^{p}	5.4	23.1^{j}	21.6^{k}	6.6	23.0^{l}	21.6^{m}	6.3	23.8^{k}	21.8^{n}	8.3
200	22.4^{k}	20.3^{m}	9.0	22.0°	20.4^{q}	7.5	–	–		21.3^{m}	20.2^{n}	5.1	22.5^{m}	20.4°	9.5

[a:] Tukey mean comparison within each oil type over temperature range (95% Confidence) no significant difference between values sharing same letter.

Table 11. Parameters *a*, *b* and *c* corresponding to the modified Andrade equation used to predict the viscosity of each vegetable oil.

Oil type	a	B	c	Temp range (°C)
Canola	−0.40	−478.44	545692.29	23–200
Corn	0.46	−804.36	560589.98	23–200
Olive	0.09	−771.31	603521.93	23–180
Peanut	0.47	−1065.63	654506.48	23–200
Soybean	1.23	−1466.38	686086.16	23–200

Table 12. Predicted dynamic viscosity values for each vegetable oil by the modified Andrade equation and their corresponding percentage error.

	Canola oil			Corn oil			Olive oil			Peanut oil			Soybean oil		
	μ_{oil} (mPa·s)			μ_{oil} (mPa·s)			μ_{oil} (mPa·s)			μ_{oil} (mPa·s)			μ_{oil} (mPa·s)		
T (°C)	EXP	PRED	E%	EXP	PRED	E%	EXP	PRED	E%	EXP	PRED	E%	EXP	PRED	E%
23 ± 1	63.5^{a}	63.5^{a}	0.0	59.2^{a}	59.5^{a}	0.5	74.1^{a}	74.3^{a}	0.2	72.2^{a}	72.4^{a}	0.3	57.1^{a}	57.1^{a}	0.1
40	34.9^{b}	34.2^{b}	2.1	34.5^{b}	33.5^{c}	3.0	40.1^{b}	39.3^{b}	1.9	38.8^{b}	38.0^{c}	2.1	31.3^{b}	31.1^{b}	0.5
60	18.8^{c}	19.4^{c}	2.7	19.1^{e}	19.8^{d}	3.8	21.1^{c}	22.0^{c}	3.9	20.5^{e}	21.1^{d}	3.0	17.9^{c}	18.0^{c}	0.5
80	11.8^{d}	12.0^{d}	2.3	12.5^{f}	12.9^{f}	3.0	13.4^{d}	13.6^{d}	1.8	12.6^{f}	13.1^{f}	3.2	11.4^{d}	11.5^{d}	1.6
100	8.2^{e}	8.1^{e}	1.3	9.0^{g}	9.0^{g}	0.3	9.6^{e}	9.1^{e}	4.9	8.8^{g}	8.7^{g}	0.6	8.3^{e}	8.0^{e}	3.2
120	5.5^{f}	5.8^{f}	4.2	6.4^{h}	6.6^{h}	2.7	6.1^{f}	6.5^{f}	6.2	6.1^{h}	6.3^{h}	2.9	5.6^{f}	5.9^{f}	5.2
140	4.4^{g}	4.3^{g}	0.9	5.1^{i}	5.1^{i}	0.2	4.8^{g}	4.9^{g}	0.4	4.8^{i}	4.7^{i}	1.4	4.8^{g}	$4.6^{g,h}$	4.2
160	$3.7^{g,h}$	$3.4^{h,i}$	7.8	4.3^{j}	4.1^{j}	4.4	$4.0^{g,h}$	$3.8^{g,h}$	5.8	4.0^{j}	3.7^{j}	7.6	$3.9^{h,i}$	$3.7^{i,j}$	4.9
180	$3.0^{h,i,j}$	$2.7^{i,j}$	9.6	3.5^{k}	3.4^{k}	2.7	3.3^{h}	3.1^{h}	7.1	3.1^{k}	3.0^{k}	2.7	$3.2^{i,j,k}$	$3.1^{j,k}$	3.4
200	$2.6^{i,j}$	2.2^{j}	14.7	$3.1^{k,l}$	2.9^{l}	8.8	—	—		2.7^{k}	2.5^{k}	7.0	2.8^{k}	2.7^{k}	5.2

[a]: Tukey mean comparison within each oil type over temperature range (95% Confidence) no significant difference between values sharing same letter.

the values of surface tension of oils at high temperature if the free fatty acid composition, density and critical parameters of the oil are known.

Viscosity

Viscosity of the oils was modeled up to 200°C using the modified Andrade equation (Eq. 10). The coefficients a, b and c were obtained using regression analysis. The Andrade equation is a form of the Arrhenius equation and uses kinematic viscosity values to establish relation between viscosity and temperature. Conversions between dynamic and kinematic viscosities were done using the following equation:

$$\mu = v\rho \tag{12}$$

where v is the kinematic viscosity (mm^2/s), μ is the dynamic viscosity ($mPa \cdot s$), and ρ the density (g/cm^3) at the selected temperature. The values of *a*, *b* and *c* were obtained using the method of least squares (Table 11). Similar trends to previous studies [18,20,22] were observed in the current study using a different assembly. According to the data, there is a high degree of accuracy between the experimental and the predicted data for all the temperatures used in the study (Table 12). The error percentage increased with increasing temperature, however the experimental and predicted data had similar trends for change in viscosity with temperature (Fig. 6).

Conclusion

Based on the results, temperature had a significant effect on all three physical properties of oil measured. Oil type was shown to have a significant effect on viscosity and density, but did not influence surface-tension. Surface tension and density decreased linearly with increasing temperature, whereas the decrease in viscosity followed a power law model. The trends for all the

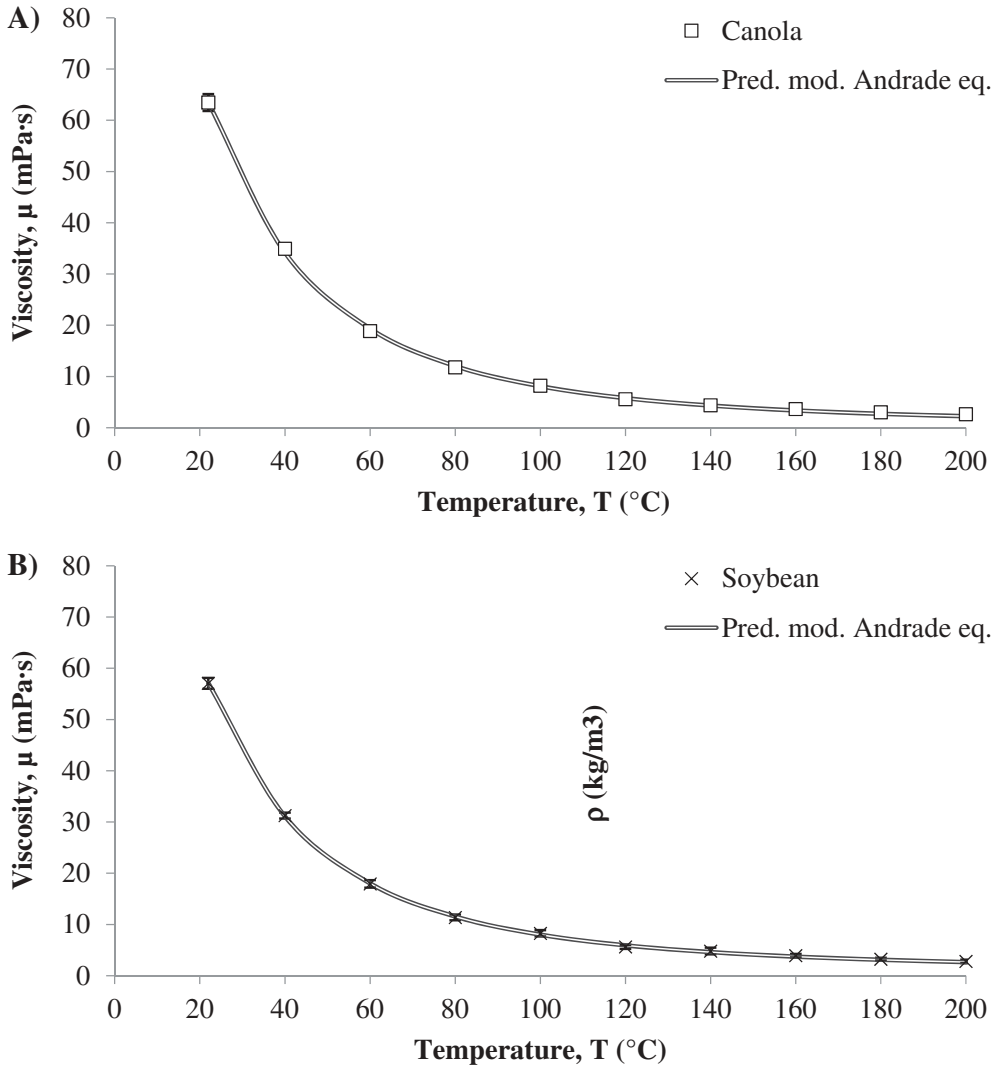


Figure 6. Comparison between viscosity experimental values of A) Canola oil, and B) Soybean oil and their corresponding predicted values by the modified Andrade equation.

three physical properties were similar to those reported in literature, and thus results could be corroborated. Mathematical models built to predict the change in surface tension (the Eötvös equation and modified Rackett-Eötvös equations), density (the modified Rackett equation), and viscosity (the modified Andrade equation) seemed to agree well with the experimental data. The error percentage for mathematical models increased with increasing temperature, however the model and experimental values followed the same trend at the range of temperatures studied. The mathematical models developed can thus be used to predict the change in physical properties of oil at high temperatures; specially for controlling process parameters during frying, spray drying, dairy processing or atomization during biodiesel production. Understanding of transport rate of oil at high temperatures (up to 200°C) when different types oils are used can be used for varied applications such as a better/more accurate prediction of the oil absorption rates during frying, and understanding of drying rate during spray drying.

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