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Thermodynamic Properties of Peanut, Canola and Rosa Mosqueta Oils

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ABSTRACT

This paper contains the results of a new experimental study of the temperature effect on density and ultrasonic velocity for peanut (Arachis hypogaea), canola (Brassica napus L., Brassica rapa L. and Brassica juncea) and rosa mosqueta (Rosa affinis rubiginosa L.) oils. The Halvorsen's model (HM), and Collision Factor Theory (CFT) were selected for prediction of these properties, attending to ease of use and range of application. An accurate response was observed, despite of the use of molecular group contribution procedures for estimation of theoretical critical points and the complex nature of the studied fluids.

Introduction

In food industry, thermodynamic properties are the most important parameters required in the design of processes. Knowledge of these magnitudes (heat capacity, density, viscosity, refractive index, phase equilibria, etc) is of practical interest to the industrial manufacture of fats and oils since thermal and mechanical procedures applied are close related on their temperature and pressure dependence. Despite of their economic importance, no systematic projects of consistent and complete compilation of its thermodynamic properties have been developed, a relative scarce of data being encountered in what is referred to these compounds. Different previous published works report data compilations of physical and thermal properties of fats and oils [1-12] but this information is not systematic, it can be found disperse and many properties have not been studied in a wide extension. The studied compounds are neither simple nor one-molecular-structured-around, their thermodynamic properties are strongly dependent of double bound presence, chain length, isomeric structures of fatty acids and molecular package of esters into solvents. An important factor that may be taken into account to understand their thermodynamic trend is the temperature influence which is strongly dependent on the freedom degrees of the molecular structure.

Among the different thermodynamic properties of solvents, volumetric and ultrasonic magnitudes have proved particularly informative in elucidating molecular interaction into liquid media, being these values of main interest for direct industrial applications. The oils studied here have in common, besides a growing economic importance, applications in food, medical or cosmetic uses and, at the same time, a severe gap in terms of physico-chemical data disposability into scientific or academic open literature.

Peanut, also known as groundnut (*Arachis hypogaea*), is a crop of global importance. It is widely grown in the tropics and subtropics, with a world annual production of about 46 million tons per year, being the biggest producers China (37% of world production) Africa (25%) and India (21%).

Peanuts have a variety of industrial end uses. Paint, varnish, lubricating oil, leather dressings, furniture polish, insecticides, and nitroglycerin are made from this kind of vegetable oil. Soap is made from saponified oil, and many cosmetics contain peanut oil and its derivatives. Peanut oil is considered as edible and often used in cooking, because it has a mild flavor and a relatively high smoke point. Due to its high monounsaturated content, it is considered health oil, showing a strong resistance to rancidity.

Canola is term to describe several varieties of the Brassicaceae family of plants, as well as, the edible oil produced from the seed of Brassica napus L., Brassica rapa L. e Brassica juncea, containing less than 2% erucic acid. This oil is considered to be among the healthiest vegetable derived oils, having a relatively low amount of saturated fat and a high content of polyunsaturated fats. This reputation has created high demand in markets around the world, and overall it is the third-most widely consumed vegetable oil. It has many non-food uses and, like soybean oil, is often used interchangeably with non-renewable petroleum-based oils in products, including industrial lubricants, biodiesel, lipsticks and inks.

Rosa mosqueta (*Rosa affinis rubiginosa L.*) is a species rose, native to Europe and West Asia, which has naturalized in America. Like most species roses, it is light pink, has five petals in single blossom form and blooms in spring or summer, although may have an occasional repeat flowering. It is from the fruit, or "hip" of this plant that bears a tiny seed that is then pressed into rose hip seed oil. The rosa mosqueta oil is widely prized for its healing, antiaging benefits, making it an oil that often is found in blends formulated for mature and sun damaged skin. It's rich in essential fatty acids that keep skin healthy, preserve its appearance by maintaining and strengthening cell membranes, and aid in overall tissue regeneration, restore skin elasticity and minimize the appearance of wrinkles like synthetic retinoic acid, but without side effects.

In the last few years a considerable effort has been developed on physico-chemical properties of organic

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chemicals but no systematic analogous projects have been developed for food technology, a relative scarce of data being encountered in oils and fats, despite their economical importance in global market. With these facts in mind, as a continuation of our scientific work investigating physical properties related to equipment design of natural oils industries [13-15], we present the temperature dependence (288.15-333.15 K) of density, and ultrasonic velocity for a collection of oils with a triacylgliceric molecule as backbone, peanut (Arachis hypogaea), canola (Brassica napus L., Brassica rapa L. and Brassica juncea) and rosa mosqueta (Rosa affinis rubiginosa L.) oils. From the experimental data, temperature dependent polynomials were fitted, the corresponding parameters being gathered.

Because of the expense of the experimental measurement of such data and current processes design is strongly computer oriented then, consideration was also given to how accurate different prediction methods work. An enormous quantity of chemicals may be found in vegetable oils (free fatty acids, phenols, peroxide, monoacylglycerols, diacylglycerols, flavonoid polyphenols, polycyclic aromatic hydrocarbons and many other complex substances). The triacylglycerol molecule is often considered the main chemical structure to develop estimative studies on thermophysical properties. The Rackett equation described by Halvorsen et al. [16-17] was tested for density estimation. This method requires the critical properties of the fatty acids and considers their composition as input. The Collision Factor Theory [18-19] was used for estimation of the ultrasonic velocity.

Attending to the obtained results, it should be concluded that the tested models offer accurate results despite geometrical simplifications and the use of estimated critical magnitudes by a group contribution method.

Materials and measurement devices

The oils (cold pressed quality), supplied by usual local providers, were stored in sun light protected form and constant humidity and temperature in our laboratory. They were analysed to determine their fatty acids compositions, the procedure being described earlier [15]. The average molar mass was computed as follows:

$$M_{oil} = 3 \cdot \left(\sum_{i=1}^{N} x_i . M_i \right) + 2 \cdot M_{CH_2} + M_{CH} \quad (1)$$

being x_i the molar fraction and M_i the molar mass of each fatty acid without a proton, N the number of fatty acid found by analysis and M_{CH_2} and M_{CH} are the molar mass contributions of glyceride molecule residue. The variation in the composition between different samples affects mainly the mono and polyunsaturated fatty acids, the change in molar mass being lower than ±1 g mol^{-1}. The molar mass and fatty acids composition are gathered in Table I.

Densities and ultrasonic velocities were measured with an Anton Paar DSA-48 vibrational tube densimeter and sound analyser, with a resolution of 10^{-5} gcm^{-3} and 1 ms^{-1}. Apparatus calibration was performed periodically in accordance with vendor instructions using Millipore quality water and ambient air at each temperature. Accuracy in the measurement temperature was better than $\pm 10^{-2}$ K by means of a temperature control device that applies the Peltier principle to maintain isothermal conditions during the measurements. Earlier works describe the experimental procedure usually applied in our laboratory [13-15].

Table I. Molar mass and fatty acids compositions of the studied oils.

Oil	Molar Mass (gmol^{-1})	Fatty Acids Composition (mass%)
PEANUT	876.16	Palmitic (16:0) 13.0 Stearic (18:0) 3.3 Oleic (18:1) 42.3 Linoleic (18:2) 37.2 Linolenic (18:3) 2.8 Araquídic (20:0) 1.4
CANOLA	882.38	Miristic (14:0) 0.1 Palmitic (16:0) 4.7 Palmitoleic (16:1) 0.3 Stearic (18:0) 1.3 Oleic (18:1) 65.3 Linoleic (18:2) 19.2 Linolenic (18:3) 8.3 Gadoleic (20:1) 0.7 Erucic (22:1) 0.1
ROSA MOSQUETA	878.26	Palmític (16:0) 5.5 Stearic (18:0) 2.0 Oleic (18:1) 15.5 Linoleic (18:2) 46.0 Linolenic (18:3) 30.0 Araquídic (20:0) 0.7 Gadoleic (20:1) 0.3

The experimental and disposable literature data of density, and ultrasonic velocity of the oils at 298.15 K [20-34] are gathered in Table II.

Table II. Experimental and literature data of density (gcm^{-3}) and ultrasonic velocity (ms^{-1}) for the studied vegetable oils at 298.15 K.

Oil	Exp. Dens.	Lit. Dens.	Exp. Ultra. Vel.	Lit. Ultra. Vel.
PEANUT	0.909411	0.913[a] (293.15 K) 0.9110-0.9250[b] (288 K) 0.903[c] 0.914[d] 0.908799[e] 0.911[f] 1.14[g] (323 K)	1448.37	N A
CANOLA	0.913505	0.9067[h] 0.9100-0.9170[i] (288 K) 0.9145[j] (293 K) 0.9129[k] (293 K) 0.913293[e] 0.87-0.90[l] 0.914-0.920[m] (293 K) 0.914-0.917[n] (293 K)	1452.00	1454.70[k]
ROSA MOSQUETA	0.922718	0.927[o] (293 K)	1455.67	N A

[a]Subrahmanyam et al., 1994
[b]Ullmann´s Encyclopedia of Industrial Chemistry, 1995
[c]Andrew et al., 2012
[i]Bailey´s Industrial oil & fat products, 2005
[j]Esteban et al., 2012
[k]Nikolic et al., 2012

[d]Musa et al., 2012
[e]Neagu et al., 2013
[f]Pandurangan et al., 2014
[g]Hussain et al., 2015
[h]Noureddini et al., 1992
[l]Zhao et al., 2014
[m]Gunstone, 2009
[n]Daun et al., 2015
[o]Concha et al., 2006

Data treatment

The measured physical properties were correlated as a function of temperature using Eq. 2:

$$P = \sum_{i=0}^{N} A_i T^i \qquad (2)$$

where P is density (gcm^{-3}), ultrasonic velocity (ms^{-1}), or isentropic compressibility (TPa^{-1}), T is absolute temperature in Kelvin and A_i are fitting parameters. N stands for the extension of the mathematical serie, optimised by means of the Bevington test. The fitting parameters were obtained by the unweighted least squared method applying a fitting Marquardt algorithm. The root mean square deviations were computed using Eq. 3, where z is the value of the property, and n_{DAT} is the number of experimental data.

$$\sigma = \left(\frac{\sum_{i=1}^{n_{DAT}} \left(z_{exp} - z_{pred} \right)^2}{n_{DAT}} \right)^{1/2} \qquad (3)$$

Fitting parameters of the Eq. 2 and the root mean square deviations are gathered in Table III. In Figures 1-3, the temperature trend of density, ultrasonic velocity and isentropic compressibility (computed by the Newton-Laplace equation from density and ultrasonic velocity) are gathered.

These figures show a diminution of density and ultrasonic velocity when temperature rises, due to a strong diminution of the packing efficiency of the triacylglycerol by molecules kinetics, as well as, a growing difficult of packing molecules by steric hindrance. At high temperatures, canola oil shows a sharp diminution of density and ultrasonic velocity when compared with other oils, showing lower values of these magnitudes than peanut oil at the highest temperature.

As expected, the isentropic compressibilities increase when temperature rises for the three oils, due to the inverse relation of this magnitude with density and ultrasonic velocity. Rosa mosqueta oil shows the highest values of the three oils studied here.

As commented, a considerable gap of disposable information was observed when open literature of volumetric or acoustic properties was looked forward. Only scarce data at single temperatures for density and only one reference of ultrasonic velocity was found for the studied oils. Table 1 gathers the disposable individual values from open literature, compared with our new experimental measurements. Figures 4 and 5 show comparison of density values for peanut and canola oils with open literature as a function of temperature. As observed, the disposable data of density of peanut oil are coincident at low temperatures, showing rising divergences at higher temperatures. It is important to highlight as the founded collections do not describe adequately the slating tendency of peanut oil density (Figure 4), sometimes showing a non-realistic behavior [20].

In what is referred to disposable data for canola oil, it was observed as the open literature data describe a linear tendency of density and, in general terms, lower values than those obtained in our experimental measurements

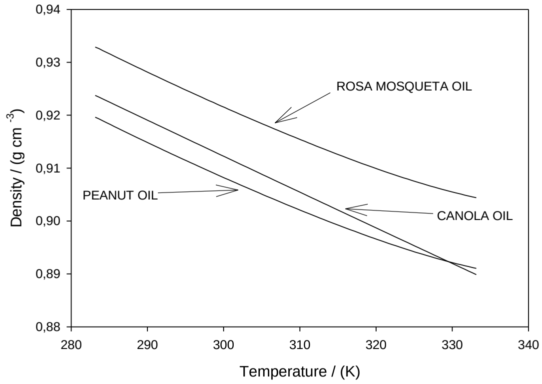

Figure 1. Temperature influence on vegetable oils density.

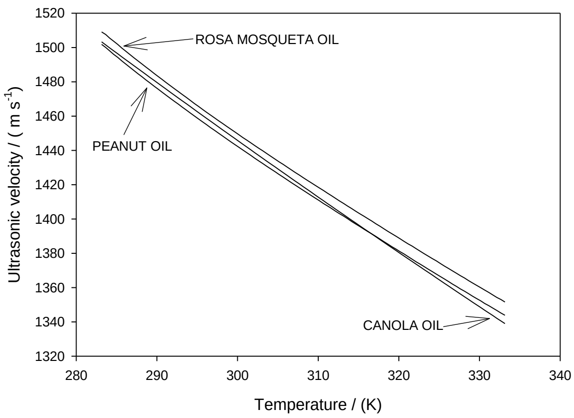

Figure 2. Temperature influence on vegetable oils ultrasonic velocity.

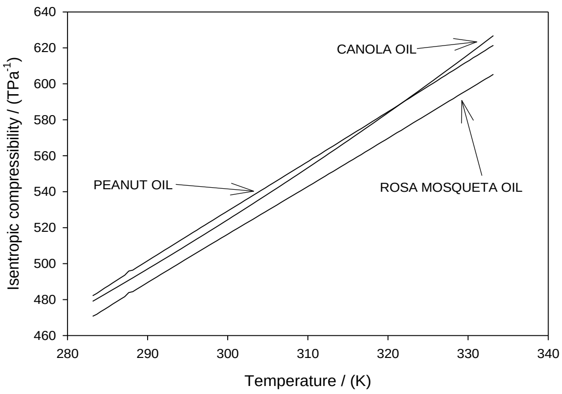

Figure 3. Temperature influence on vegetable oils isentropic compressibility.

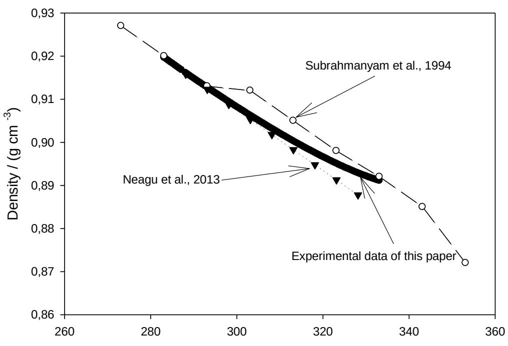

Figure 4. Comparison of experimental peanut oil density data with open literature data.

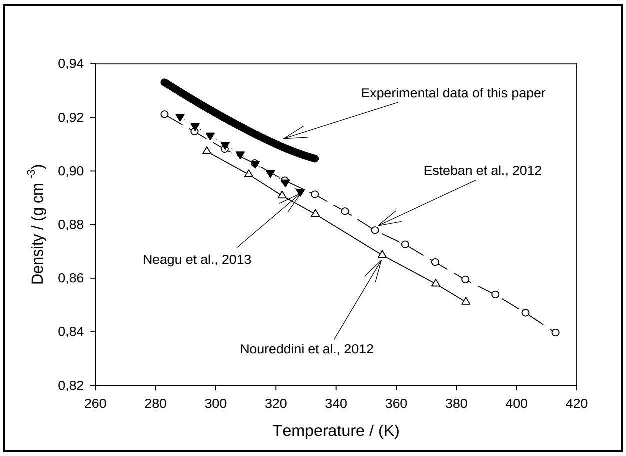

Figure 5. Comparison of experimental canola oil density data with open literature data.

Results and discussion

Critical point prediction

Constantinou and Gani [35] developed an advanced group contribution method for critical point estimation, based on the UNIFAC molecular groups. This procedure allows a second order level of contributions, overcoming the limitation of traditional group contribution models which cannot distinguish isomers or resonance structures into the studied compounds. This method is quite reliable for all critical properties, though there can be significant errors for some smaller substances due to group additivity is not so accurate for small molecules even though it may be possible to form them from available groups.

This method was applied to obtain the critical point of the fatty acids, and then used into the prediction methods that will be indicated above. The observed deviations when compared with database information [36] are really low.

Table IV gathers the critical points for the enclosed fatty acids into the studied vegetable oils.

Prediction of densities

The physical property packages used in chemical simulators typically rely on generalized equations for predicting properties as a function of temperature, pressure, etc.

Table IV.Estimated critical properties for the enclosed fatty acids into the studied vegetable oils by Constantinou and Gani method [35].

Fatty acids	P_c(MPa)	T_c(K)	Z_c	ω
Myristic (14:0)	0.1651	762.51	0.2165	0.7184
Palmitic (16:0)	0.1431	780.38	0.2076	0.8007
Palmitoleic (16:1)	0.1462	781.32	0.2083	0.7891
Oleic (18:1)	0.1280	797.50	0.1999	0.8699
Linoleic (18:2)	0.1306	798.36	0.2006	0.8585
Linolenic (18:3)	0.1333	799.20	0.2013	0.8470
Stearic (18:0)	0.1255	796.65	0.1993	0.8813
Arachidic (20:0)	0.1113	811.57	0.1917	0.9601
Gadoleic (20:1)	0.1134	812.36	0.1923	0.9489
Erucic (22:1)	0.1013	826.09	0.1853	1.0261

Despite the success developing several procedures of density estimation for pure compounds or mixtures, really, only a few of them may be of practical application for fats and oils. One proposed correlation that holds promise for application to oils is the Rackett equation of state. The modification of this equation by Halvorsen et al. [16-17] has demonstrated to be accurate, only requiring critical magnitudes for the enclosed fatty acids. If these magnitudes are not known, they must be estimated as indicated. The method of Halvorsen is described as follows:

$$\rho = \frac{\sum x_i \cdot M_i}{R \cdot \left(\frac{\sum x_i \cdot T_{ci}}{P_{ci}} \right) \cdot \left(\sum x_i \cdot \beta_i \right)^{[1+(1-T_r)^{2/7}]}} + F_C \qquad (4)$$

where ρ is the oil density, x_i is the mole fraction of fatty acids into that oil, M_i is the molar mass of each fatty acid, R is the universal constant of gases, P_{ci} is the critical pressure of each fatty acid and T_r is the reduced temperature. The β parameter is the compressibility factor for the original equation of Rackett (Zc) or an acentric factor dependent parameter if we use the modified Rackett equation (Z_{RA}) [37]. The mixing rule to compute the pseudocritical temperature, and then the reduced temperature of the oil is described as follows:

Table III. Parameters of Eq. 2 for 283.15-333.15 K and the corresponding root mean square deviations (Eq 3).

$\rho/(gcm^{-3})$					
	A_0	A_1	A_2	A_3	σ
PEANUT	2.328066 10^{-1}	8.770940 10^{-3}	-3.380947 10^{-5}	4.025864 10^{-8}	3.07 10^{-5}
CANOLA	1.126157	-7.334201 10^{-4}	2.657605 10^{-8}	1.379430 10^{-10}	1.43 10^{-6}
ROSA MOSQUETA	1.987113 10^{-1}	9.219854 10^{-3}	-3.522617 10^{-5}	4.174946 10^{-8}	3.26 10^{-5}
$u/(ms^{-1})$					
	A_0	A_1	A_2	A_3	σ
PEANUT	7.803619 10^{3}	-5.309901 10^{1}	1.527893 10^{-1}	-1.549122 10^{-4}	9.52$10^{-2}$
CANOLA	3.053206 10^{3}	-8.221474	1.236657 10^{-2}	-9.405726 10^{-6}	9.94 10^{-2}
ROSA MOSQUETA	7.934687 10^{3}	-5.423934 10^{1}	1.562524 10^{-1}	-1.583681 10^{-4}	2.09 10^{-1}
$\kappa_S/(TPa^{-1})$					
	A_0	A_1	A_2	A_3	σ
PEANUT	-1.947039 10^{3}	1.893351 10^{1}	-5.287520 10^{-2}	5.759342 10^{-5}	7.78 10^{-2}
CANOLA	-1.825647 10^{2}	3.718329	-1.060411 10^{-2}	2.021651 10^{-5}	7.78 10^{-3}
ROSA MOSQUETA	-1.920072 10^{3}	1.863530 10^{1}	-5.192225 10^{-2}	5.625301 10^{-5}	1.41 10^{-1}

$$T_r = \frac{T}{\sum x_i T_{ci}} \quad (5)$$

F_c is a correction factor proposed by Halvorsen which depends on the oil structure backbone. The correction factor equation for the studied is:

$$F_c = 0.0236 + 0.000082 \cdot (875 - M_{oil}) \quad (6)$$

where M_{oil} is the molar mass of each studied oil, as gathered into Table I. Table V shows the root square deviations for predictive density values by Halvorsen's model (HM) versus experimental data at 298.15 K.

Table V. Root mean square deviations (g/cm^3) for Halvorsen method density prediction for the studied vegetable oils.

PEANUT	1.0899^{-2}
CANOLA	1.6424^{-2}
ROSA MOSQUETA	2.8330^{-2}

Prediction of ultrasonic velocities

In the last few years an increasingly interest for the application of low/high frequency ultrasound techniques for thermodynamic applications has occurred [38]. Ultrasonic velocity has been systematically measured but this kind of data is scarce yet. In terms of fats and oils, ultrasonic measurements are extremely rare. The experimental data were compared with the values obtained by the Collision Factor Theory (CFT) [18-19], which is dependent on the collision factors among molecules as a function of temperature:

$$u = \frac{u_\infty \cdot \left(\sum_{i=1}^{3} (x_i \cdot S_i) \right) \cdot \left(\sum_{i=1}^{3} (x_i \cdot B_i) \right)}{V} \quad (8)$$

where u_∞ is 1600 m/s, S_i is the collision factor of each fatty acid, B_i is the molecular volume of each fatty acid and V is the molar volume considering each oil a mixture of fatty acids attending to the composition (Table I).

The collision factors (S) were estimated using open literature for fatty acids density [36] and Wada method for estimation of ultrasonic velocity of each fatty acid [39]. The deviations for CFT method are gathered in Table VI.

Table VI. Root mean square deviations (m/s) for CFT ultrasonic velocity prediction for the studied vegetable oils at 298.15 K.

PEANUT	226.39
CANOLA	212.66
ROSA MOSQUETA	183.97

Conclusions

This paper contains the results of a new experimental study of the effect of temperature on density, and ultrasonic velocity for peanut (Arachis hypogaea), canola (Brassica napus L., Brassica rapa L. and Brassica juncea) and rosa mosqueta (Rosa affinis rubiginosa L.) oils, due to their rising economic importance in terms of food technology, personal-care developments and traditional medicinal uses. The measured experimental data contributes for a better characterization of these vegetable oils and increase the disposable data for theoretical works and modeling of macromolecules. Consideration was also given to how accurate different prediction methods work, due to increasing importance of theoretical procedures in simulation and process design.

The tested methods (Halvorsen's model (HM), and the Collision Factor Theory (CFT)) showed accurate capability of prediction at the range of application, despite of different simplifying assumptions, the use of estimated critical magnitudes by molecular group contribution approach and the complex nature of the studied fluids.

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