

# Temperature Influence on Thermodynamic Properties of Argan (*Argania Spinosa*), Neem (*Azadirachta Indica*) and Common Walnut (*Juglans Regia L.*) Oils

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**Abstract**—This paper contains the results of a new experimental study of the influence of temperature on density, refractive index and ultrasonic velocity for argan (*Argania spinosa*), neem (*Azadirachta indica*) and common walnut (*Juglans regia L.*) oils, due to their rising economic importance in terms of food technology, personal-care products, as well as traditional medicinal uses. Consideration was also given to how accurate different prediction methods work, due to the key role of theoretical procedures in simulation and process design. The Halvorsen's model (HM), Gharagheizi model (GM) and the Collision Factor Theory (CFT) were selected for prediction, attending to ease of use, accuracy and range of application. An accurate response was observed, despite of the use of estimated critical magnitudes by molecular group contribution approach and the complex nature of the studied fluids.

**Keywords**—Density, Refractive index, Ultrasonic velocity, Vegetable oil, Temperature, Modeling

## I. INTRODUCTION

Vegetable oils extracted from nuts, seeds, or fruits are important in food technology and many other industrial sectors as pharmaceutical, personal-care products, bioenergy, lubricants, etc. During the process of extraction, purification and final use, the oils undergo different physical and chemical modifications, then accurate knowledge of thermo physical properties of these fluids is key in terms of quality control analysis and design equipments and processes of oil processing. In recent years, different types of oils have gained prominence primarily for its cosmetic qualities and beneficial properties for skin and body care applications. Although traditionally used, in the last few years their economic importance increases dramatically [1-3].

In this work, we gather different properties as a function of temperature of three oils extracted from argan (*Argania spinosa*), neem (*Azadirachta indica*) and common walnut (*Juglans regia L.*). The oils studied here are of regional culture (argan mainly in Morocco, neem in India and Burma but extended to other tropical and subtropical countries, and

common walnut spread from the Mediterranean to China) but with growing importance in the global economy of oils and fats.

Argan oil is produced from the kernels of the argan tree (*Argania spinosa*) that is endemic to Morocco. The fruit of the argan tree is small, and round, oval, or conical. A thick peel covers the fleshy pulp, surrounding a hard-shelled nut (approximately 25% of the fresh fruit). This nut contains one to three argan oil-rich kernels. (2) Extraction yields of the oil in the kernels are from 30% to 50%, depending on the method applied. To extract the kernels, workers first dry argan fruit and then, remove the fleshy pulp. Different attempts to mechanize this process have been unsuccessful, so workers still do it hardly by hand, as traditionally always it has been done. Argan oil has become increasingly popular for cosmetic use and as additive for personal-care preparations. The number of products commercially available on the global market with this oil or derivatives as ingredient increased to over 100 in the last few years. The increasing market penetration of argan oil has prompted the Moroccan government to increase production until 4,000 tonnes by 2020.

Neem oil has an extensive history of use in India for a variety of therapeutic purposes. Neem oil is not used for cooking but it offers a wide spectrum of potential uses for preparing cosmetics (soap, hair products, body and hand creams, etc) and in folklore traditional medicine, as well as, in the treatment of different afflictions. Neem oil is a vegetable oil pressed from the fruits and seeds of the neem (*Azadirachta indica*), an endemic evergreen tree in the Indian subcontinent that has been introduced to many other areas around the world in the tropic areas. Neem oil varies in color from golden yellow to bright red. It is composed mainly of triglycerides and contains many triterpenoid compounds. Azadirachtin is the most well known triterpenoid in neem oil. The azadirachtin content of neem oil varies from 300 ppm to over 2500 ppm depending on the extraction technology and

quality of the neem seeds crushed. Nimbin is another triterpenoid which has been credited with some of neem oil's properties as an antiseptic, antifungal, antipyretic and antihistaminic. Neem oil also contains many types of sterols (campesterol, beta-sitosterol, stigmasterol, and many others). A common walnut is the nut of any tree of the genus *Juglans* (Family Juglandaceae), particularly the Persian or English walnut, *Juglans regia*. Technically a walnut is the seed of a drupaceous nut, and thus not a true botanical nut. It is used for nutritional applications after being processed while green for pickled walnuts or after full ripening for its nutmeat. Nutmeat of the eastern black walnut from the *Juglans nigra* is less commercially available, as are butternut nutmeats from *Juglans cinerea*. The walnut is nutrient-dense with protein and many essential fatty acids. Walnut oil is also known to be a remedy to treat fungal infections and psoriasis.

The worldwide production of walnuts has been increasing rapidly in the last few years, with the largest increase coming from Asia. The world produced a total of 2.55 million metric tonnes of walnuts in 2010; China was the world's largest producer of walnuts. The other major producers of walnuts were Iran, United States, Turkey, Ukraine, Mexico, Romania, India, France and Chile.

In the last few years a considerable effort has been developed on physico-chemical properties of organic 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. Among the different thermodynamic properties of solvents, the volumetric, optical and ultrasonic properties have proved particularly informative in elucidating molecular interaction into liquid media. They are of practical interest into industrial manufacture of oils since applied thermal and mechanical procedures are close related on thermophysical properties dependence with temperature and pressure.

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.

Continuing our scientific work investigating physical properties related to equipment design of oil industries [4-6], we present the temperature dependence (288.15-333.15 K) of density, refractive index and ultrasonic velocity for argan (*Argania spinosa*), neem (*Azadirachta indica*) and common walnut (*Juglans regia* L.) oils. From the experimental data, temperature dependent polynomials were fitted, the corresponding parameters being gathered.

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. [7-8] was tested for density estimation. This method requires the critical properties of the fatty acids and considers their

composition as input. Gharagheizi's group contribution model was applied for refractive index estimation [9]. The Collision Factor Theory [10-11] 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.

## II. EXPERIMENTAL

### A. Materials and measurement devices

The oils, 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 [6]. The average molar mass was computed as follows:

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

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  $\pm 1 \text{ g mol}^{-1}$ . The molar mass and fatty acids composition are gathered in Table I.

TABLE I: Molar mass and fatty acids compositions of the studied oils

Oil	Molar Mass (g mol <sup>-1</sup> )	Fatty Acids Composition (mass%)
ARGAN	886.52	Oleic (18:1) 43.0–49.0 Stearic (18:0) 4.0–7.0 Linolenic (18:3) < 0.2 Linoleic (18:2) 29.0–36.0 Palmitoleic (16:1) 0.3–3.0 Behênic (22:0) < 0.2 Palmitic (16:0) 11–15 Arachidic (20:0) < 0.5 Miristic (14:0) < 0.1
NEEM	869.91	Miristic (14:0) 2.6 Palmitic (16:0) 13.6–14.9 Stearic (18:0) 14.4–19.1 Oleic (18:1) 49.1–61.9 Linoleic (18:2) 7.5–15.8
WALNUT	879.11	Palmitic (16:0) 6.0–8.0 Stearic (18:0) 1.0–3.7 Araquidic (20:0) < 0.2 Oleic (18:1) 14.0–23.1 Linoleic (18:2) 50.0–65.0 Linolenic (18:3) 9.0–15.0

Densities and ultrasonic velocities were measured with an Anton Paar DSA-48 vibrational tube densimeter and sound analyser, with a resolution of  $10^{-5} \text{ g cm}^{-3}$  and  $1 \text{ ms}^{-1}$ . 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  $\pm 10^{-2} \text{ K}$  by means of a temperature control device that applies the Peltier principle to maintain isothermal conditions during the

measurements. Refractive indices were measured with a Mettler RE50 refractometer with an uncertainty of  $\pm 0.00005$ , and temperature was controlled as described for the densimeter and sound analyser. Earlier works describe the experimental procedure usually applied in our laboratory [4-6].

The experimental and disposable literature data of density, refractive index and ultrasonic velocity of the oils at 298.15 K [12-25] are gathered in Table II.

TABLE II: Experimental and literature data of density ( $\text{gcm}^{-3}$ ), refractive index and ultrasonic velocity ( $\text{ms}^{-1}$ ) at 298.15 K for the studied vegetable oils

Oil	Exp. Dens.	Lit. Dens.	Exp. Refrac. Index	Lit. Refrac. Index	Exp. Ultra. Vel.	Lit. Ultra. Vel.
ARGAN	0.9107 5	0.906- 0.919 <sup>a</sup> (293 K) 0.906- 0.919 <sup>b</sup> (288 K)	1.4689 8	1.463- 1.472 <sup>a</sup> 1.4630- 1.4708 <sup>b</sup>	1446.5 3	1449 .5 <sup>c</sup> 1448 .30 <sup>d</sup>
NEEM	0.9446 1	0.912- 0.965 <sup>e</sup> 0.778 <sup>f</sup> (308 K) 0.925- 0.940 <sup>g</sup> (288 K) 0.8758 <sup>h</sup> (30 3 K) 1.024 <sup>i</sup> 0.9185 <sup>j</sup> (303 K)	1.4768 9	1.47 <sup>i</sup>	1460.9 3	1443 <sup>j</sup> (303 K)
WALNUT	0.9181 6	0.924- 0.926 <sup>k</sup> (293 K) 0.945- 0.970 <sup>l</sup> 0.945- 0.970 <sup>m</sup>	1.4745 9	1.475- 1.476 <sup>k</sup> (293 K) 1.4777- 1.4788 <sup>m</sup> 1.4730 <sup>n</sup>	1452.3 4	NA

<sup>a</sup> Charrouf and Guillaume, 2008

<sup>b</sup> Firestone, 2006

<sup>c</sup> Malaoui et al., 2005

<sup>d</sup> Malaoui, 2016

<sup>e</sup> Karmakar et al., 2012

<sup>f</sup> Ali et al., 2013

<sup>g</sup> Muthu et al., 2010

<sup>h</sup> Karunanithi and Maria, 2015

<sup>i</sup> Radha and Manikandan, 2011

<sup>j</sup> Mohammad Ali et al., 2016

<sup>k</sup> Karleskind, 1992

<sup>l</sup> Leahu et al., 2016

<sup>m</sup> Patraş and Dorobanţu, 2010

<sup>n</sup> Salunkhe and Kadam, 1995

## B. Data treatment

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

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

where  $P$  is density ( $\text{gcm}^{-3}$ ), refractive index, ultrasonic velocity ( $\text{ms}^{-1}$ ), isentropic compressibility ( $\text{TPa}^{-1}$ ),  $T$  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  $n_{\text{DAT}}$  is the number of experimental data.

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

Fitting parameters of the Eq. 2 and the root mean square deviations (Eq. 3) are gathered in Table III. In Figures 1-4, the temperature trend of density, refractive index, 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, refraction 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 due to the steric hindrance. As expected, for the three oils the isentropic compressibility increases when temperature rises, due to the inverse relation of this magnitude with density and ultrasonic velocity. Neem oil shows the highest values for these properties (density, refractive index and ultrasonic velocity), gathering below 298.15 K solid condensation that prevents density and sonic measurements.

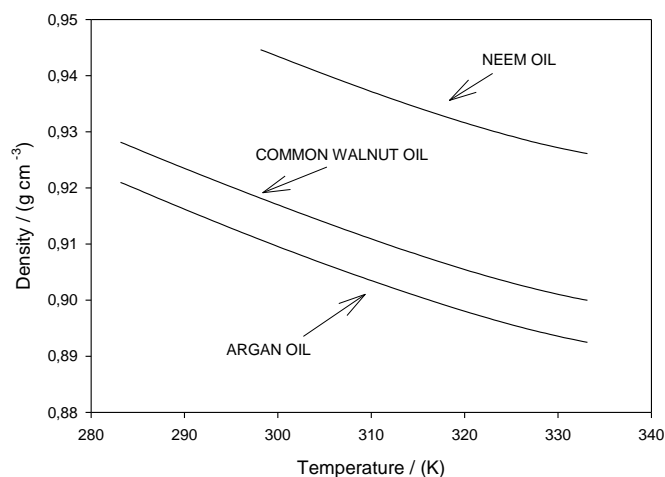


Figure 1 Temperature influence on vegetable oils density

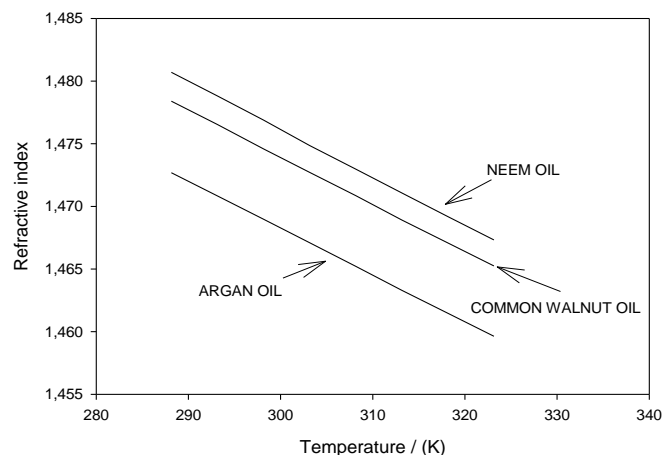


Figure 2 Temperature influence on vegetable oils refractive index

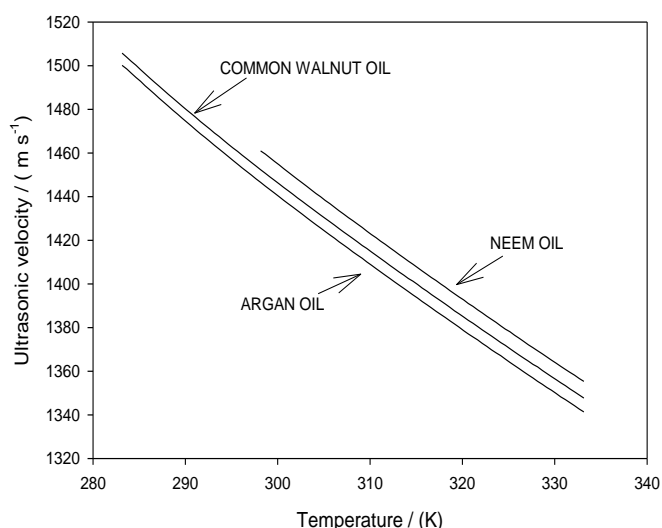


Figure 3 Temperature influence on vegetable oils ultrasonic velocity

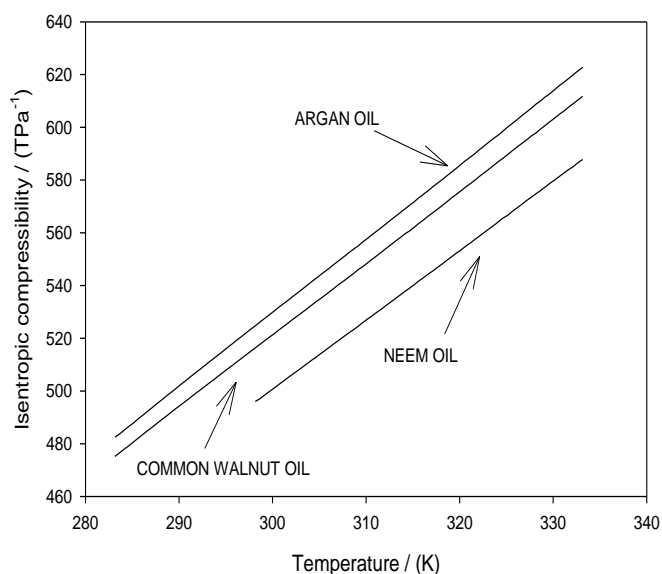


Figure 4 Temperature influence on vegetable oils isentropic compressibility

TABLE III: Parameters of Eq. 2 for the range of temperature 283.15-333.15 K and the corresponding root mean square deviations in accordance with Eq. 3.

$\rho/(\text{gcm}^{-3})$					
	$A_0$	$A_1$	$A_2$	$A_3$	$\sigma$
ARGAN	0.4026	0.0071	$-2.85 \cdot 10^{-5}$	$3.45 \cdot 10^{-8}$	$3.07 \cdot 10^{-5}$
NEEM	-0.8150	0.0192	$-6.73 \cdot 10^{-5}$	$7.61 \cdot 10^{-8}$	$1.18 \cdot 10^{-5}$
WALNUT	0.0462	0.0106	$-3.96 \cdot 10^{-5}$	$4.63 \cdot 10^{-8}$	$2.77 \cdot 10^{-5}$
$n_D$					
	$A_0$	$A_1$	$A_2$	$A_3$	$\sigma$
ARGAN	1.1054	$4.31 \cdot 10^{-3}$	$-1.53 \cdot 10^{-5}$	$1.68 \cdot 10^{-8}$	$1.08 \cdot 10^{-5}$
NEEM	1.1251	$4.27 \cdot 10^{-3}$	$-1.55 \cdot 10^{-5}$	$1.72 \cdot 10^{-8}$	$2.59 \cdot 10^{-5}$
WALNUT	1.3225	$2.28 \cdot 10^{-3}$	$-8.89 \cdot 10^{-6}$	$9.90 \cdot 10^{-9}$	$1.02 \cdot 10^{-5}$
$u/(\text{ms}^{-1})$					
	$A_0$	$A_1$	$A_2$	$A_3$	$\sigma$
ARGAN	7864.8388	-53.6264	$0.15 \cdot 10^{-1}$	$-2.00 \cdot 10^{-5}$	$9.51 \cdot 10^{-2}$
NEEM	4973.8411	-25.6193	$6.44 \cdot 10^{-2}$	$-6.04 \cdot 10^{-5}$	$6.78 \cdot 10^{-2}$
WALNUT	7692.3879	-51.9977	$1.49 \cdot 10^{-1}$	$-2.00 \cdot 10^{-4}$	$1.09 \cdot 10^{-1}$
$\kappa_S/(\text{TPa}^{-1})$					
	$A_0$	$A_1$	$A_2$	$A_3$	$\sigma$
ARGAN	-2081.6833	20.2123	-0.0570	$6.20 \cdot 10^{-5}$	$6.32 \cdot 10^{-2}$
NEEM	621.8183	-5.8202	0.0261	$-2.69 \cdot 10^{-5}$	$2.88 \cdot 10^{-2}$
WALNUT	-1742.3963	17.0033	-0.0469	$5.13 \cdot 10^{-5}$	$7.25 \cdot 10^{-2}$

### III. RESULTS AND DISCUSSION

#### A. Critical point prediction

Constantinou and Gani [26] 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. This method is quite reliable for all critical properties, though there can be significant errors for some smaller substances due to group additivity it 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 [27] are really negligible.

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



TABLE IV: Estimated critical properties for the enclosed fatty acids into the studied vegetable oils by Constantinou and Gani method [26]

Fatty acids	P <sub>c</sub> (MPa)	T <sub>c</sub> (K)	Z <sub>c</sub>	ω
Palmitic	1.4307	780.38	0.2076	0.8007
Palmitoleic	1.4617	781.32	0.2083	0.7891
Oleic	1.2802	797.50	0.1999	0.8699
Linoleic	1.3059	798.36	0.2006	0.8585
Linolenic	1.3325	799.20	0.2013	0.8470
Stearic	1.2553	796.65	0.1993	0.8813
Arachidic	1.1133	811.57	0.1917	0.9601
Miristic	1.6510	762.51	0.2165	0.7184
Behenic	0.9968	825.36	0.1848	1.0370

### B. 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. Despite the success developing several procedures of density estimation for pure compounds or mixtures, only a few of them may be of real 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. [7-8] 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:

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

where  $\rho$  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  $\beta$  parameter is the compressibility factor for the original equation of Rackett ( $Z_c$ ) or an acentric factor dependent parameter if we use the modified Rackett equation ( $Z_{RA}$ ) [28]. The mixing rule to compute the pseudocritical temperature, and then the reduced temperature of the oil is described as follows:

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

$F_c$  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 density predictions by Halvorsen's model (HM) versus experimental data at different temperatures.

TABLE V: Deviations (g/cm<sup>3</sup>) for Halvorsen method density prediction for the studied vegetable oils at 288.15, 298.15 and 333.15 K

T (K)	Argan Oil	Neem Oil	Walnut Oil
288.15	0.0343	-	0.0314
298.15	0.0340	0.0465	0.0314
333.15	0.0385	0.0520	0.0366

### C. Prediction of refractive index

The refractive index is a measure of the change in velocity of a specific light wave as it travels from one medium to another and it is directly related to the molecular structure of the material. It is frequently used to characterize organic compounds and quality control measurements.

Gharagheizi's group contribution model was applied for refractive index estimation [9]. This model is, until now, used in a larger database to compute the interaction contributions of the chemical substructures, showing a more robust trend than any other previously tested [29-33].

Based in 80 chemical structure contributions, the model computes the refractive index using the following equation:

$$n_D = \sum_{i=1}^{80} (n_i \cdot n_{Di}) + n_{D0} \quad (7)$$

where  $n_{D0}$  and  $n_{Di}$  are the intercept of the equation, the contribution of the  $i$ th chemical substructure to the refractive index of the compound, and  $n_i$  is the number of occurrences of the  $i$ th chemical substructure in every chemical structure of the pure compound, respectively. Table VI shows the root square deviations for predictive refractive index values by Gharagheizi's model (GM) versus experimental data at 298.15 K.

TABLE VI: Deviations for Gharagheizi's model (GM) refractive index prediction for the studied vegetable oils at 298.15 K

Argan oil	0.0218
Neem oil	0.0374
Walnut oil	0.2281

### D. 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. 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) [10-11], 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  $u_{\infty}$  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, considered a theoretical mixture of fatty acids attending to the composition (Table I).

The collision factors ( $S$ ) were estimated using open literature for fatty acids density [27] and Wada method for estimation of ultrasonic velocity of each fatty acid [34]. The deviations for CFT method are gathered in Table VII.

TABLE VII: Deviations ( $\text{ms}^{-1}$ ) for CFT ultrasonic velocity prediction for the studied vegetable oils at 298.15 K

Argan oil	177.5
Neem oil	336.2
Walnut oil	309.7

#### IV. CONCLUSIONS

This paper contains the results of a new experimental study of the effect of temperature on density, refractive index and ultrasonic velocity for argan (*Argania spinosa*), neem (*Azadirachta indica*) and common walnut (*Juglans regia* L.) oils, due to their rising economic importance in terms of food technology, personal-care products, as well as traditional medicinal uses. 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), Gharagheizi's model (GM) and the Collision Factor Theory (CFT)) showed accurate capability of prediction at the range of application, despite of assumptions, the use of estimated critical magnitudes by molecular group contribution approach and the complex nature of the studied fluids.

The measured experimental data contributes for a better characterization of these emerging vegetable oils and increase the disposable data for theoretical works and modeling of macromolecules

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