Temperature Effect in the Babassu (*Orbignya speciosa*) Oil: A Physico-Chemical Study

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Babassu (*Orbignya speciosa*) is a Brazilian palm with extraordinary importance in socioeconomic and ecologic terms. It is found in humid tropical areas, especially in degraded landscapes. There are several uses for babassu coconut and babassu oil. However, their immense potential for large-scale providing other industrial products still needs to be explored due to the necessity for modern scale planning and deep knowledge of vast spectrum thermodynamic properties. This paper gathers a new experimental physico-chemical study of the temperature effect on two critical properties, density and ultrasonic velocity for babassu oil, due to its rising economic significance and a high potential for intensive farming in regions with low economic resources. We consider how accurately different theoretical prediction methods work due to modern processes, design, and algorithm simulations being strongly computer-oriented. The Agrawal-Thodos equation for density and Collision Factor Theory for ultrasonic velocity was selected, mainly attending to ease of use and range of application. We observed a good response at the studied conditions, despite geometrical simplifications into triglyceride molecules and using estimated critical magnitudes by molecular group contribution approach. A broad comparison was made with disposable open literature thermodynamic data, showing an essential dispersion of data, and highlighting the quality of the experimental data presented in this work.
Keywords: Babassu Oil. Density. Ultrasonic Velocity. Prediction. Theoretical Models. Molecular Group Contribution Approach. Surrogate Oil.

Introduction

The babassu coconut palm is of Brazilian origin, found in the Amazon basin and nearby humid tropical regions, especially in degraded landscapes. It is a typical transition plant from the region of the Cerrado biome, the Amazon rainforest, and the Brazilian semiarid northeast. The *Arecaceae* family records approximately 240 genera and 2,700 species worldwide [1-4], existing 146 species in different Brazilian ecosystems, standing out in terms of economic, social, and ecological potential. Mainly species of the genera *Attalea*, *Orbygnia*, *Syagrus*, *Acrocomia*, and *Mauritia*, are often sold at popular markets in Brazil [5-8] for the traditional use of fruit and heart of palm in fresh food or processed as sweets, and drinks, oils, and crafts. In the *Orbygnia* genus, the babassu (*Orbignya speciosa*) stands out, an isolated robust palm tree of 10-30 meters high and 30-60 centimeters in diameter, with 7 to 22 pinnate leaves, measuring 4-8 meters in length [9].

Babassu (Babaçu in Portuguese) is one of the Brazilian palm trees with greater significance in terms of ecology, and society, with economic relevance for the Brazilian states of Bahia, Maranhão, Piauí, and Tocantins [10]. It shows excellent relevance for the subsistence of many traditional communities since the whole plant can be used. The leaves are used to cover traditional houses, in crafts, the stipe as a building element of the civil structure, the fruits as a source of energy (coal), almonds as food, and the manufacture of cosmetics [7-8,10-12]. The palm tree produces coconuts arranged in different separated clusters. Four main structures characterize these fruits: epicarp, mesocarp, endocarp, and internal almonds. The epicarp is formed by resistant fibers, mainly in manufacturing brushes and carpets. The mesocarp contains 20%-25% starch and is used in foods such as flour and a drink similar to chocolate. The endocarp is a raw material for the

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manufacture of insulators and the production of different materials such as methanol, acetic acid, tar, and coal. The internal almonds are inside the endocarp, and more than 60% of the almond is oil. The residual pie, after oil extraction, is usually applied for final compost and supplementary animal feed [13-18].

The fatty acid profile of babassu oil has been evaluated by different researchers[19-23], demonstrating the high content of acid lauric. The oil from babassu coconut is rich in this fatty acid, with a concentration above 40% [24,25]. The lauric fats, like babassu oil, are essential in the fats and oils industry. They are resistant to non-enzymatic oxidation, and unlike other saturated fats, they show low melting temperatures. So, depending on their physical properties and resistance to oxidation, the food industry uses them to prepare special fats for ice cream, margarine, and cocoa butter substitutes. They are also used in the cosmetics industry.

Coconuts and palm kernel oils are the principal sources of lauric fats. These fats' primary sources in Brazil are coconut, palm kernel, and babassu. In the extraction of babassu coconut oil, firstly it is used a cold pressing procedure to reduce the oil content in the almond pie up to 12-15%. Then, the dry residue is extracted by solvent, leaving a residual oil content in the solid phase lesser than 1%. The presence of lipase enzymes in vegetable oils is the catalyst for enzymatic hydrolysis and, then, rancor taste, mainly when the unsaturated fatty acids profile is high, which is not in this case. In Brazil, babassu oil is used almost exclusively in cleaning and personal-care products. Its use in food technology is still secondary, appearing only in margarine production. There is, however, a rising interest in developing new markets for the use of babassu. Nevertheless, its potential for providing other industrial products remains unexploited due to the lack of scale, adequate production structure, and deep scientific investigation [26-30].

In present times where global warming poses a severe threat, alternate sources for attending ever increasing energy demands are strongly necessary. Green fuels, non-dependent on non-sustainable wellsprings such as biodiesel and bioethanol, are promising, and efforts have been made to develop and optimize technology production and blends. Biodiesel production is one such method that is an excellent alternative source of energy. The principal concern about using edible vegetable oils for biodiesel production is that they might add problems in terms of already occurring food shortages and price levels. The solution should be on diverse sources of biodiesel production as non-edible oil and fats and waste oil from food processing. Compared to petrodiesels, biodiesel has poor low-temperature properties, low oxidative stability, high density, and lower compressibility. Alternative vegetable oils such as babassu is a non-edible oil with low cost. It contains significant amounts of shorter chain saturated acids into triglyceride, gathers low free fatty acid value, and shows a higher cetane number with better oxidative stability.

Thermodynamic properties are the most critical parameters required in equipment design and processes. Physico-chemical properties of biodiesel or petrodiesel blends, such as density, ultrasonic velocity, and compressibility, depend on the dissimilar chemical composition of the fuels. These magnitudes influence combustion, quality of gas emission, and injection timing [31-34]. Density is one of the most crucial properties of the fuel due to pumps and injectors must deliver an amount of finely adjusted fuel precisely for adequate internal combustion [35]. Compressibility defines spray characteristics upon injection. Since biodiesel's compressibility is lower than petrodiesel, the injection timing can cause different gas emissions performances [36]. The fuel injection process occurs almost in the isentropic condition in the combustion device. Then, isentropic compressibility is the most appropriate magnitude to estimate the fuel injection timing. The experimental procedure to obtain direct isentropic compressibility measurements is acoustic. Ultrasonic velocity measurements are simple experimental procedures with accurate results and wide acceptance for any blends [37-40].

Concerning the physical properties related to oils and fats industry equipment design [41-46], we present the temperature dependence (288.15-323.15 K) of density and ultrasonic velocity for babassu oil (*Orbignya speciosa*). We fitted the temperature-dependent polynomials from the experimental data, and the corresponding parameters were gathered. The current process design is strongly computer-oriented, so we considered how accurately different prediction methods work.

It is possible to find many chemicals in vegetable oils, such as free fatty acids, phenols, peroxide, monoacylglycerols, diacylglycerols, flavonoid polyphenols, polycyclic aromatic hydrocarbons, and many other complex substances. However, the triacylglycerol molecule is often considered the main chemical structure of vegetable oils. Therefore, to develop calculations using mathematical models, we consider a surrogate oil simulating that a single triglyceride molecule can adequately describe babassu oil with a fatty acid content proportional to its analytic composition (Table 1).

The Agrawal-Thodos (AT) equation [47] for density and the Collision Factor Theory (CFT) procedure [48] for ultrasonic velocity were selected for prediction, attending to ease of use and range of application. A good response at the studied conditions was observed, despite geometrical simplifications into triglyceride molecules and using estimated critical magnitudes by Joback's molecular group contribution approach [49]. An exhaustive comparison was made with disposable open literature thermodynamic data, showing an essential dispersion of data, and highlighting the quality of the experimental data presented in this work.

Materials and Methods

Materials and Measurement Devices

The oil, supplied by usual local providers (Cocal Maranhense, Itapecuru-Mirim, Maranhão-Brazil), was stored in sunlight-protected form and constant humidity and temperature in our laboratory. The provider analyzed it to determine its fatty acid compositions and the standard applied procedure described earlier [50]. The average molar mass was computed as follows:

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

being x_i molar fraction and M_i the molar mass of each fatty acid without a proton, N the number of fatty acids found by analysis and M_{CH2} and M_{CH} are the molar mass contributions of glycerin molecule residue. The variation in the composition

Table 1. Molar mass and fatty acids composition of babassu oil.

Compound	Molar Mass ($gmol^{-1}$)	Fatty Acids Composition (mass%)
Babassu oil (*Orbignya speciosa*)	676.278	Caprylic (8:0) 4.74 Capric (10:0) 5.17 Lauric (12:0) 50.46 Myristic (14:0) 13.91 Palmitic (16:0) 9.70 Stearic (18:0) 8.94 Oleic (18:1) 5.16 Linoleic (18:2) 1.92

between different samples mainly affects mono and polyunsaturated fatty acids, the change in molar mass being lower than ±1 g mol^{-1}. Table 1 shows the molar mass and fatty acids composition of babassu oil. The obtained results are similar to those observed in the disposable open literature [21,51].

Densities and ultrasonic velocities were measured by an Anton Paar DSA-5000M vibrational tube densimeter and sound analyzer, with a resolution of 10^{-5} gcm^{-3} and 1 ms^{-1}. Apparatus calibration was performed periodically by vendor instructions using Millipore quality water and ambient air at each temperature. Accuracy in the measurement temperature was better than ±10^{-2} K. The obtained measurements were coincident, in general terms, with earlier published data (Table 2) [51-79].

Data Treatment

The measured physical properties were correlated as a function of temperature using the Equation 2:

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

where P is density (gcm^{-3}) or ultrasonic velocity (ms^{-1}), T is the absolute temperature in Kelvin, and A_i is fitting parameters. N is the extension of the mathematical series optimized using the Bevington test. The unweighted least squared method obtained the fitting parameters by applying a fitting Marquardt algorithm. The root means square deviations were computed using the Equation 3, where z is the property's value, and nDAT 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 Equation 2 and the root mean square deviations, attending to Equation 3, are gathered in Table 3. Figures 1 and 2 present the temperature trend of density,

Table 2. Experimental/literature density and ultrasonic velocity for babassu oil at 298.15 K.

Compound	ρ/(gcm^{-3})		u/(ms^{-1})	
	Exptl.	Lit.	Exptl.	Lit.
Babassu oil (*Orbignya speciosa*)	0.91886	0.9140 [53] 0.9047 [54] 0.923 [55] 0.920 [56] 0.9151 [57] 0.929 [61] 0.9202-0.9204 [62] 0.9169 [63] 0.956 [66] 0.914-0.917 [67] 0.923 [68] 0.9137-0.9171 [70] 0.9122-0.9152 [78]	1411.35	NA

NA: Non Available.

ultrasonic velocity, and isentropic compressibility, computed by means Newton-Laplace equation from density and ultrasonic velocity experimental measurements. These figures show a progressive diminution of density and ultrasonic velocity when the temperature rises due to a sharp diminution of packing efficiency of the enclosed macromolecules into the bulk phase. Both combined effects, higher molecular kinetics by rising temperatures and steric hindrance of heavy molecular structures, produce a growing difficulty in packing molecules. Figure 1b shows a zoom of density trend into Figure 1a around the 288.15-298.15 K range, in which a higher number of earlier literature data were found. Figure 2 presents the inverse relation of isentropic compressibility and density and ultrasonic velocity by Laplace-Newton equation, showing an inverse relationship with temperature.

Results and Discussion

Isobaric Compressibility

A frequently applied derived quantity is the temperature dependence of volumetric properties, which is expressed as isobaric expansibility or thermal expansion coefficient (α), obtained from accurate isobaric experimental data of volumetric trend at a specific range of temperature. The data reported in the literature usually show only values of the thermal expansion coefficients of pure compounds and their mixtures, showing the relative changes in density, calculated by ($-\Delta\rho/\rho$) as a function of temperature, and assuming that a remains constant over the temperature range. In the case of pure chemicals, these oils can be computed by the Equation 4:

$$\alpha = -\left(\frac{\partial \ln \rho}{\partial T}\right)_{P,x} \tag{4}$$

taking into account the strong temperature dependence of density (Figure 1a). Attending to this relation, Figure 3 shows the isobaric expansibility of babassu (*Orbignya speciosa*) oil. Only two previously published collections of isobaric expansibilities were found. As observed, our experimental values gather decreasing negative values for rising temperatures, showing a similar trend to Barañano and colleagues (2019) [52] but different from those data obtained from Ceriani and colleagues (2008) [73].

Critical Point Prediction

The basis for the design and theoretical estimation work into chemical processes is a collection of data related to the thermodynamic trend of the compounds evolved. However, many times is not possible to find open literature with reasonable values of properties compounds. So, theoretical estimation methods are generally employed. Group-contribution methods have been used, and in the last few years, a vast collection

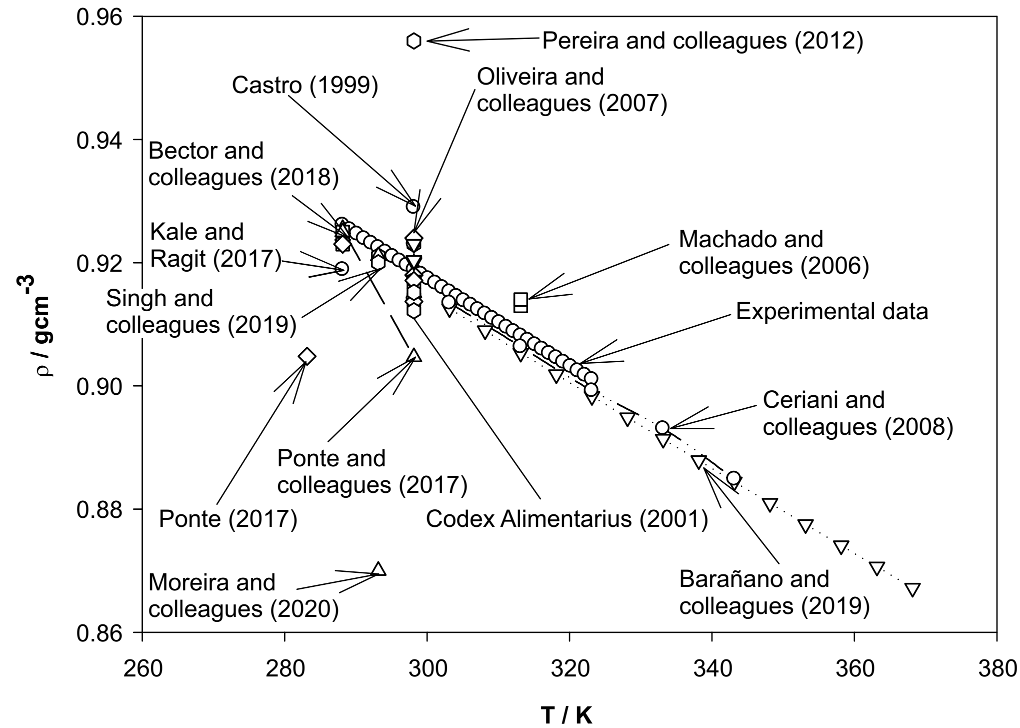
Table 3. Fitting parameters of Equation 2 for 288.15-323.15 K and the corresponding root mean square deviations (Equation 3).

$\rho/(gcm^{-3})$					
Compound	**A0**	**A1**	**A2**	**A3**	**σ**
Babassu oil	1.257755E+00	-1.853207E-03	3.410485E-06	-3.377978E-09	5.203964E-06

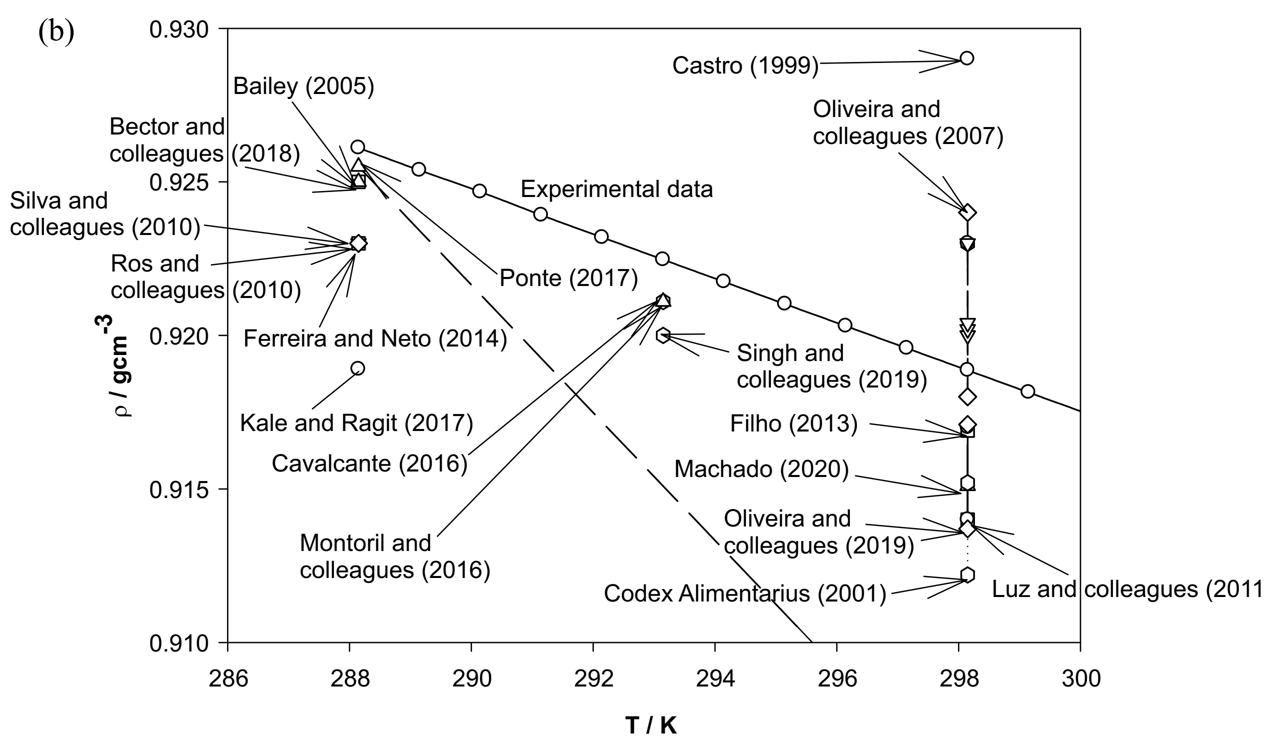
$u/(ms^{-1})$					
Compound	**A0**	**A1**	**A2**	**A3**	**σ**
Babassu oil	1285.76	8.971444	-0.044655	53.586930E-06	6.062544E-02]

Figure 1. (a) Density values, (b) zoom of density values figure, and (c) ultrasonic velocity for babassu oil at a range of temperature (288.15-323.15 K).

(a)



(b)



(c)

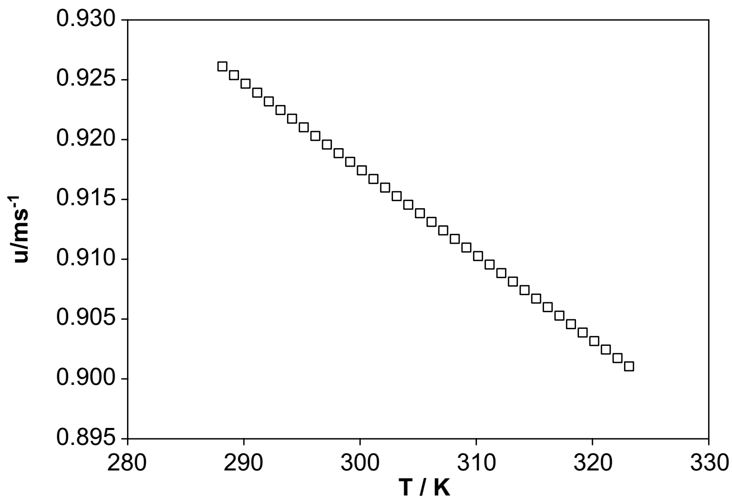


Figure 2. Isentropic compressibility (TPa^{-1}) for babassu oil at a range of temperature (288.15-323.15 K).

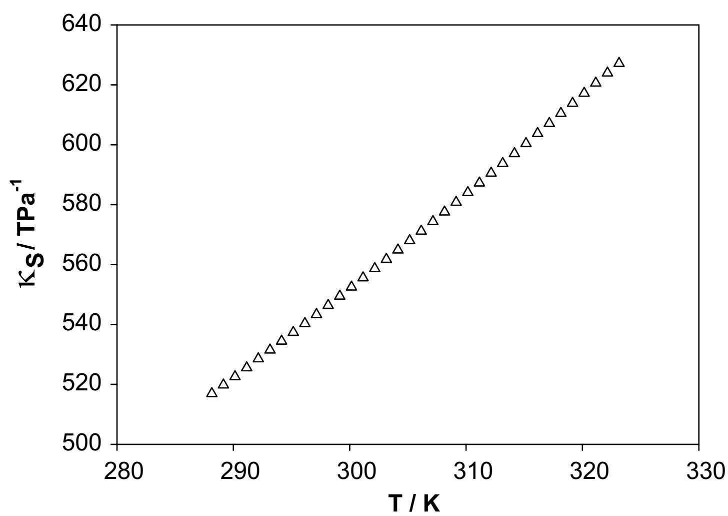
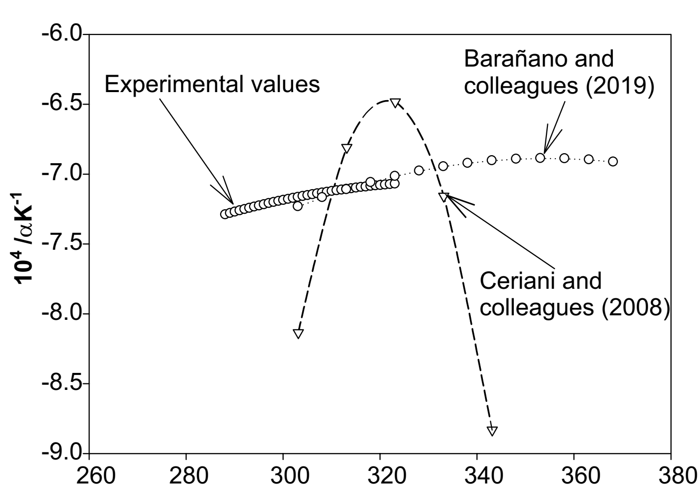


Figure 3. Isobaric expansibility (K^{-1}) for babassu oil at a range of temperature (288.15-323.15 K).



of methods have been proposed [49,80]. In these methods, the property of a compound is a function of different structurally-dependent parameters, which are determined by summing the frequency of each molecular group enclosed into the molecule and its particular contribution. Marrero-Gani [81] developed an advanced group contribution method for critical point estimation of covalent molecules based on the UNIFAC molecular group conception. The application of three different sets of functional groups, one for a first-order approximation and the other two sets for refining the estimations for complex compounds, providing additional structural information, has led to an advanced and robust group-contribution method for the estimation of acute thermodynamic properties. It overcomes the limitation of traditional group contribution models that cannot adequately distinguish isomers or resonance structures. This method was applied to obtain the critical point of the surrogate triglyceride of babassu oil for use in density estimation. Compared with database information on different oils, the observed deviations are negligible. Table 4 gathers the computed critical point for the studied oil.

Prediction of Densities

The physical property packages used in chemical simulators typically rely on generalized equations for predicting properties as a function of temperature, and pressure, among others. Despite the success of developing several procedures of density estimation for pure compounds or mixtures, only a few of them may be of the actual application for fats and oils. One proposed correlation that holds promise for application to oils is the Agrawal-Thodos (AT) (Equation 5) [47]:

$$\rho = 1 + k \cdot \left(1 - \frac{T}{T_C}\right)^{\beta} \quad (5)$$

where T and T_C are work temperature and critical temperature, respectively. This relationship requires that parameters k and β be established to define the density of a saturated liquid for temperatures extending up to the critical point of this fluid. The k and β parameters were proposed in the original paper. Initially, this equation was developed for simple covalent cryogenic fluids. So, this model should be applied because babassu oil is a surrogate oil composed of a unique triglyceride molecule (Table 1). Table 5 presents the deviations of this model gathered at the studied temperature range.

Prediction of Ultrasonic Velocities

There has been an increasing interest in low/high-frequency ultrasound techniques for industrial engineering in the last few decades. Then, ultrasonic velocity has been systematically measured. However, the literature needs to be more comprehensive regarding the range of work conditions or complex mixtures such as natural fats and oils. Moreover, the data about substances' ultrasonic measurements are insufficient in databases. The DIPPR database (DIPPR, Design Institute for Physical Properties) is an extensive physical data source on chemical compounds. It gathers rigorously evaluated results of experimental measurements and high-accuracy correlations for computation as functions of the temperature at saturated conditions. However, despite this extensive compilation of high-quality data,

Table 4. Critical properties for surrogate triglyceride molecule of babassu oil by Marrero-Gani group contribution method [81].

Compound	**Pc (bar)**	**Tc (K)**	**Vc ($cm^3 mol^{-1}$)**
Babassu oil (*Orbignya speciosa*)	8.139	965.962	2436.212

Table 5. Root mean square deviations (gcm^{-3}) for AT density prediction for babassu oil at the studied temperature range.

Compound	k	β	σ
Babassu oil (*Orbignya phalerata Mart.*)	-10.80012E-3	-5.511988E+0	3.663758E-4

ultrasonic velocity needs to be gathered among the variety of thermodynamic magnitudes enclosed in this database. In the last few years, a necessity for ultrasonic data was noticed in the open literature, mainly for complex mixtures and biological nature compounds. The Collision Factor Theory (CFT) [48] computes the isentropic compressibility using collision factors parameters, which are a function of temperature in pure solvent or mixture. The Equation 6 could express this model:

$$\kappa_S = \rho^{-1} \cdot \left(\frac{u_\infty \cdot S \cdot B}{V} \right)^{-2} \quad (6)$$

where u_∞ is 1600 m/s, S is the collision factor, B is the actual molecule volume per mole, and V is the molar volume. The collision factors (S) of the pure solvents (the fatty acids as explained in Table 1) used in the CFT calculations were estimated by using the ultrasonic velocities of each fatty acid using Wada's group contribution method previously proposed by Freitas and colleagues 2013) [82]. The characteristic molar volumes were calculated by the group contribution method of Bondi. The experimental data for the babassu oil studied here were compared with the obtained values as surrogate oil by CFT procedure. Table 6 gathered the deviation for ultrasonic velocity estimation by the CFT method.

Comparison with Open Literature Data

In the last decades, it has boosted the use of natural oils, different from those usually applied for vast food stock production, searching for low cost, green procedures, and circularity in new business opportunities. Despite the commercial interest of these oils, the open scientific literature contains few physicochemical data, which are essential for the design of equipment and processes. Valuable collections of accurate data and information as a function of temperature into a wide range are scarce for complex fluids such as oils. It is often not easy to identify the final quality of the data because the purification process of the solvents, the device calibration, or the accuracy of the measurements needs to be commented upon in original published papers. Only a few collections of density data for the studied oil are disposable, as earlier commented [51-79]. As expected, information related to ultrasonic measurements is mainly more dispersed and scarce. As far as we know, no disposable data for this oil are available for ultrasonic velocity. Figures 1a and 1b show a comparison between our experimental measurements and that data from open literature as a function of temperature. Relative agreement is verified for a few collections of density data [52, 60, 65, 69, 77] observed substantial deviations for many works containing data on this oil.

Table 6. Collision factor, molecular volume (cm3), and percentage error for CFT ultrasonic velocity prediction for babassu oil at 298.15K.

Compound	S (cm^{-3})	B	% Error
Babassu oil (*Orbignya phalerata Mart.*)	1.0850	218.88	7.602

Conclusions

Babassu is one of the Brazilian palm trees with greater significance in terms of ecology, society, and economic relevance. The fatty acid profile of babassu oil has been evaluated by different researchers, demonstrating the high content of acid lauric, being these lauric fats of high importance for the fats/oils industry with core applications in the cosmetic, food industry, and confectionery. However, despite their interest and rising new markets for the use of babassu, its potential for providing other industrial products remains unexploited due to the lack of mainly adequate scientific characterization and physico-chemical investigation, as explained earlier [51-79].

This work gathers an extensive collection of density and ultrasonic velocity at the temperature range 288.15-323.15 K. Prediction methods for density and ultrasonic velocity worked well in terms of accuracy, despite introduced approximations and group contribution procedures used to analyze critical points of evolved fatty acids. A broad comparison was made with disposable open literature thermodynamic data, showing a vital dispersion of values. As previously commented, disposable open literature offers only a few collections of density data for babassu oil and invalid previously measured data for ultrasonic velocity. Although, in general terms, a relative agreement is observed for a few collections of density data [52, 60, 65, 69, 77] observed solid deviations for many works containing data on this oil, highlighting the quality of the experimental data presented in this work.

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