Viscosities and Densities of Binary Mixtures of Coconut + Colza and Coconut + Soybean Biodiesel at Various Temperatures

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Viscosity η and density ρ of binary mixtures of coconut + colza and coconut + soybean biodiesel have been measured over the whole composition range at T = (293.15, 313.15, 333.15, 353.15, and 373.15) K and at atmospheric pressure. Densities and viscosities data were correlated with R-square adjustment and the Andrade equation, respectively, with good agreement. The deviations of viscosities ($\Delta\eta$) have been calculated from the experimental data. To predict the density and viscosity we have used Joback's method and a group contribution model proposed by Ceriani et al. (*J. Chem. Eng. Data* **2008**, *53*, 877−880), respectively.

Introduction

Renewable energy has gained attention during the last few decades because of the decreasing oil supply and increasing environmental consciousness. The production of biodiesel from vegetable oils is one of the alternatives for a world strategy to expand renewable energy sources. Several methods exist for producing biodiesel. The most significant is the transesterification method using a basic homogeneous catalyst. Biodiesel has many advantages,[1] and it has been increasingly used as a substitute of conventional diesel fuel. It may be directly used in diesel vehicles with little or no modification.[2] Nowadays, in Brazil, the Brazilian Regulatory Agency (Agência Nacional do Petróleo, Gás Natural e Biocombustíveis, ANP)[3] established a biodiesel content of B4 (in other words, 4 % of biodiesel) on the diesel blends commercialized. This value will reach 5 % in 2013.

The knowledge of thermodynamic properties of biodiesel (and their blends) plays an important role in the understanding of intermolecular interactions between the different molecules of biodiesel produced by vegetable oils used in its production, which affect their thermodynamic properties. Viscosities and densities data are also important in numerous chemical engineering unit operations, being also an input for process simulation and optimization to model the combustion processes and other applications. We believe that there is still a lack of systematic studies on data of thermodynamics property of biodiesel blends and biodiesel−diesel over the whole composition range at different operational conditions of pressure and temperature.[4−11] For this reason, in an attempt to improve the available viscosity and density data bank in the literature, here, we have reported the viscosity η and density ρ at atmospheric pressure and at T = (293.15, 313.15, 333.15, 353.15, and 373.15) K for binary mixtures of coconut + colza and coconut + soybean biodiesel over the whole composition range.

Experimental Section

Biodiesel oil samples used in this study were obtained from commercial vegetable oils. Coconut oil was gently furnished by Petrobras S/A. Commercial samples of colza and soybean oils were bought from a local supplier in the city of Fortaleza (Ceará/Brazil). The biodiesel samples were obtained by alkaline-catalyzed (potassium methoxide, 100 w = 8) transesterification of coconut, colza, and soybean oils using methanol (supplied from J. T. Baker) as transesterificant alcohol, without any pretreatment to the feedstocks due to their lower acidity number. Methanol in excess, about volume fraction (100 φ) = 100, was used in those reactions. Transesterification reactions were carried out homogeneously in the liquid phase. The reaction procedure consisted of an initial addition of the oil and alcohol phases, followed by the catalyst loading under mechanical agitation (1250 rpm), at 298 K during 1 h in a laboratory scale apparatus. After the transesterification, the resulting glycerin was removed by decantation after 12 h, and the resulting ester phase was washed in three steps. The first step was accomplished with a twice-washing procedure with pure water (100 φ = 10) to remove catalyst, soap, and glycerol excess. After that, it was washed once with chloridric acid (0.1 M), 100 φ = 10, to neutralize the medium. It should be noticed that after each washing procedure the dense phase was separated by decantation.

All biodiesel samples were characterized following laboratory procedures and standards indicated by the Brazilian Regulatory Agency (ANP),[3] before blends preparation. All blends were prepared in the mass fraction range between 100 w = (10 and 90), at 298.15 K.

The compositions of the esters were analyzed by gas chromatography (GC) using a Varian CP-3800 gas chromatograph system equipped with a flame ionization detector (FID; T_{detector} = 523.15 K) and automated split injector ($T_{\text{injection}}$ = 473.15). The column was a CP WAX 52CB 30 m × 0.25 mm × 0.05 μm DB (T_{column} = 483.15 K).

At the beginning of each measurement a volume of 5 mL of sample was filled into the Anton Paar SVM 3000 digital oscillation U-tube apparatus to measure the densities ρ and

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Table 1. Physicochemistry Properties of Pure Biodiesel of Coconut, Soybean, and Colza

physical property	coconut biodiesel	colza biodiesel	soybean biodiesel	limits[3]		
				USA ASTM D 6751	EU EN 14214	Brazil ANP42
flash point/K	386.15	> 423.15	437.15	403.15 min	393.15 min	373.15 min
ester content/w	98.2	97.8	99.1		96.5 min	96.5
free glycerin content/w	0.013	0.020	0.004	0.02 max	0.02 max	0.02 max
total glycerin content/w	0.072	0.157	0.154	0.24 max	0.25 max	0.38 max
cold filter plugging point/K	268.15	256.15	264.15		5 max (grade A) 0 max (grade B) −5 max (grade C) −10 max (grade D) −15 max (grade E) −20 max (grade F)	
acidity number/(mg KOH$\cdot$g^{-1})	0.0320	0.1025	0.1145	0.5 max	0.5 max	0.5 max
kinematic viscosity, at 313.15 K/(mm$^2\cdot$s^{-1})	2.45	4.47	3.96	1.9 to 6.0	3.5 to 5.0	3.00 to 6.00
density/(kg$\cdot$m^{-3})	871.9	883.8	884.3		860 to 900	850 to 950

dynamic viscosities η of pure biodiesels and their binary mixtures. The measuring cell contains a tube filled with sample, which rotates at constant speed. To calculate the kinematic viscosity from the measured dynamic viscosity, the density must be known. For this reason, the SVM 3000 also has a density cell using the well-known oscillating U-tube principle. Both the density cell and the viscosity cell are filled in one single procedure; the two different measurements occur simultaneously. The binary mixtures for the measurements of densities and viscosities were prepared by mass using an electronic balance (Technal Mark 210 A) accurate to 0.1 mg. Density measurements have an uncertainty of ± 0.0005 g$\cdot$cm^{-3} and viscosity a relative uncertainty of ± 0.35 %. The temperature in the cell was regulated to ± 0.01 K, with an uncertainty of ± 0.02 K. The uncertainty in mole fractions reported on this work is estimated to be less than $\pm\ 1.8\cdot10^{-3}$.

Prediction. For viscosity prediction the method proposed by Ceriani et al.[7] was used. This method uses the well-known concept of group contribution, in which a compound or a mixture of compounds is considered as a solution of groups and its properties are the sum of the contribution of each group. The equations for this model are

$$\begin{aligned}\ln(\eta_i) = {} & \sum_k N_k\left(A_{1k} + \frac{B_{1k}}{T} - C_{1k}\ln T - D_{1k}T\right) + M\sum_k N_k \\ & \left(A_{2k} + \frac{B_{2k}}{T} - C_{2k}\ln T - D_{2k}T\right) + Q \qquad (1)\end{aligned}$$

$$Q = \xi_1 q + \xi_2 \qquad (2)$$

$$q = \alpha - \frac{\beta}{T} - \gamma\ln T - \delta T \qquad (3)$$

$$\xi_1 = f_0 - N_c f_1 \qquad (4)$$

$$\xi_2 = s_0 - N_{cs} s_1 \qquad (5)$$

where η_i is the dynamic viscosity, in mPa$\cdot$s; T is the temperature, in K; N_k is the number of groups k in the molecule i; M is the component molecular weight that multiplies the "perturbation term"; A_{1k}, A_{2k}, B_{1k}, B_{2k}, C_{1k}, C_{2k}, D_{1k}, and D_{2k} are parameters obtained for the regression of data experimental by Ceriani et al.;[7] Q is a correction term; ξ_1 and ξ_2 are related to each class of compounds, where ξ_1 is a function of N_C, the total number of carbon atoms in the molecule, and ξ_2 describes the differences between the vapor pressure of isomer esters at the same temperature and is related to the number of carbons of the substitutive fraction N_{CS}, in the fatty esters, which is mainly used to account for the effect of the alcoholic potion. α, β, γ, δ, f_0, f_1, s_0, and s_1 are the optimization parameters.

To estimate the critical properties (T_c, P_c, and V_c) and normal boiling temperature (T_b) of biodiesel we have used the Joback method of group contributions and the Lee−Kesler mixing rules, which is recommended by Knapp et al.[12] In Joback method, the properties of each constituent of biodiesel were calculated. After that, the critical properties of biodiesel were computed by Lee−Kesler equations.

Table 2. Fatty Acid Mass Fraction w of Vegetable Oil Methyl Esters

fatty acid methyl ester	coconut 100 w	soybean 100 w	colza 100 w
C 8:0	4.08		
C 10:0	3.65		
C 12:0	35.35		
C 14:0	19.84		
C 16:0	13.83	11.32	3.99
C 18:0	3.94		3.91
C 18:1	14.30	25.68	56.67
C 18:2	4.73	54.94	23.61
C 18:3		8.07	9.88
C 20:0			1.94

Table 3. Experimental Densities (ρ) and Viscosities (η) for Coconut, Colza, and Soybean Biodiesel at the Temperature Range T = (293.15 to 373.15) K

biodiesel	T/K	ρ/g$\cdot$cm^{-3}	η/mPa$\cdot$s
coconut	293.15	0.8709	3.8417
	313.15	0.8555	2.4574
	333.15	0.8402	1.7226
	353.15	0.8246	1.2816
	373.15	0.8093	0.9880
colza	293.15	0.8846	7.4296
	313.15	0.8701	4.4727
	333.15	0.8556	2.9959
	353.15	0.8412	2.1577
	373.15	0.8268	1.6294
soybean	293.15	0.8853	6.4440
	313.15	0.8707	3.9640
	333.15	0.8562	2.6979
	353.15	0.8416	1.9659
	373.15	0.8272	1.4959

Results and Discussion

Table 1 summarizes some physicochemistry properties (flash point, ester content, free glycerin content, total glycerin content, cold filter plugging point, acidity number, kinematic viscosity, and density) of the pure biodiesel samples of coconut, colza, and soybean, along with Brazilian, European, and American specifications.

The compositions of esters were determined by GC-FID. The fatty acid composition of methyl esters are presented in Table 2.

For the coconut + colza and coconut + soybean systems we have conducted viscosity and density measurements of nine and

Table 4. Dynamic Viscosity, η, Viscosity Deviation, $\Delta\eta$, and Density, ρ, for the Binary Mixtures (w_1 Coconut Biodiesel + (1 − w_1) Colza Biodiesel), at Different Temperatures

	η/mPa·s		$\Delta\eta$/mPa·s		ρ/g·cm^{-3}	
w_1	T/K = 293.15	T/K = 313.15	T/K = 293.15	T/K = 313.15	T/K = 293.15	T/K = 313.15
0.0962	6.9443	4.2070	–0.1401	–0.0718	0.8831	0.8683
0.1967	6.4856	3.9544	–0.2383	–0.1219	0.8819	0.8672
0.2989	6.0532	3.7165	–0.3040	–0.1538	0.8803	0.8655
0.3970	5.6734	3.5026	–0.3318	–0.1700	0.8792	0.8643
0.4974	5.3235	3.3037	–0.3215	–0.1666	0.8779	0.8630
0.5977	4.9710	3.1065	–0.3141	–0.1617	0.8764	0.8614
0.6952	4.6708	2.9382	–0.2645	–0.1335	0.8756	0.8605
0.8039	4.3549	2.7537	–0.1904	–0.0989	0.8740	0.8587
0.9017	4.0908	2.6028	–0.1036	–0.0527	0.8726	0.8572
	T/K = 333.15	T/K = 353.15	T/K = 333.15	T/K = 353.15	T/K = 333.15	T/K = 353.15
0.0962	2.8311	2.0450	–0.0423	–0.0284	0.8540	0.8395
0.1967	2.6754	1.9405	–0.0700	–0.0449	0.8526	0.8379
0.2989	2.5279	1.8411	–0.0874	–0.0547	0.8509	0.8362
0.3970	2.3938	1.7494	–0.0966	–0.0605	0.8495	0.8347
0.4974	2.2682	1.6642	–0.0944	–0.0577	0.8481	0.8332
0.5977	2.1411	1.5767	–0.0937	–0.0574	0.8464	0.8314
0.6952	2.0350	1.5015	–0.0757	–0.0471	0.8452	0.8300
0.8039	1.9162	1.4174	–0.0561	–0.0360	0.8433	0.8280
0.9017	1.8163	1.3477	–0.0315	–0.0200	0.8419	0.8265

	η/mPa·s	$\Delta\eta$/mPa·s	ρ/g·cm^{-3}
w_1	T/K = 373.15	T/K = 373.15	T/K = 373.15
0.0962	1.5482	–0.0195	0.8250
0.1967	1.4734	–0.0298	0.8233
0.2989	1.4015	–0.0362	0.8215
0.3970	1.3351	–0.0396	0.8199
0.4974	1.2728	–0.0375	0.8184
0.5977	1.2090	–0.0370	0.8165
0.6952	1.1536	–0.0299	0.8149
0.8039	1.0912	–0.0225	0.8129
0.9017	1.0389	–0.0121	0.8112

Table 5. Dynamic Viscosity, η (mPa·s), Viscosity Deviation, $\Delta\eta$ (mPa·s), and Density, ρ (g·cm^{-3}), for the Binary Mixtures (w_1 Coconut Biodiesel + (1 – w_1) Soybean Biodiesel), at Different Temperatures

	η/mPa·s		$\Delta\eta$/mPa·s		ρ/g·cm^{-3}	
w_1	T/K = 293.15	T/K = 313.15	T/K = 293.15	T/K = 313.15	T/K = 293.15	T/K = 313.15
0.0983	4.0344	2.5743	–0.0631	–0.0312	0.8726	0.8572
0.1979	4.2860	2.7169	–0.0707	–0.0387	0.8741	0.8588
0.3060	4.4109	2.7935	–0.1248	–0.0657	0.8757	0.8605
0.4016	4.7310	2.9782	–0.1514	–0.0817	0.8772	0.8620
0.6002	5.2614	3.2806	–0.1422	–0.0811	0.8800	0.8650
0.8041	5.8526	3.6216	–0.0816	–0.0473	0.8829	0.8681
0.9057	6.1854	3.8122	–0.0132	–0.0097	0.8841	0.8695
	T/K = 333.15	T/K = 353.15	T/K = 333.15	T/K = 353.15	T/K = 333.15	T/K = 333.15
0.0983	1.8010	1.3382	–0.0175	–0.0107	0.8419	0.8264
0.1979	1.8927	1.4022	–0.0229	–0.0148	0.8457	0.8284
0.3060	1.9463	1.4419	–0.0364	–0.0222	0.8452	0.8299
0.4016	2.0620	1.5251	–0.0506	–0.0302	0.8469	0.8319
0.6002	2.2573	1.6570	–0.0507	–0.0353	0.8501	0.8352
0.8041	2.4780	1.8123	–0.0288	–0.0195	0.8534	0.8357
0.9057	2.6000	1.8978	–0.0059	–0.0036	0.8550	0.8404

	η/mPa·s	$\Delta\eta$/mPa·s	ρ/g·cm^{-3}
w_1	T/K = 373.15	T/K = 373.15	T/K = 373.15
0.0983	1.0323	–0.0056	0.8111
0.1979	1.0802	–0.0083	0.8130
0.3060	1.1109	–0.0125	0.8148
0.4016	1.1722	–0.0189	0.8168
0.6002	1.2680	–0.0248	0.8202
0.8041	1.3836	–0.0128	0.8240
0.9057	1.4464	–0.0016	0.8259

seven intermediate compositions, respectively, (plus two for the pure components) at atmospheric pressure. Biodiesel samples measurements start at the temperatures of (293.15 to 373.15) K, in steps of 20 K. The results of our measurements (dynamic viscosity, density, and viscosity deviation) are shown in Tables 3, 4, and 5. It is important to mention that these values are an average of at least three concurrent measurements. The density values obtained on this work were compared with literature data[4,6,11] showing good agreement, with relative errors less than 0.5 %.

For the purpose of visualization, some data from the aforementioned tables are depicted in Figure 1 that illustrate

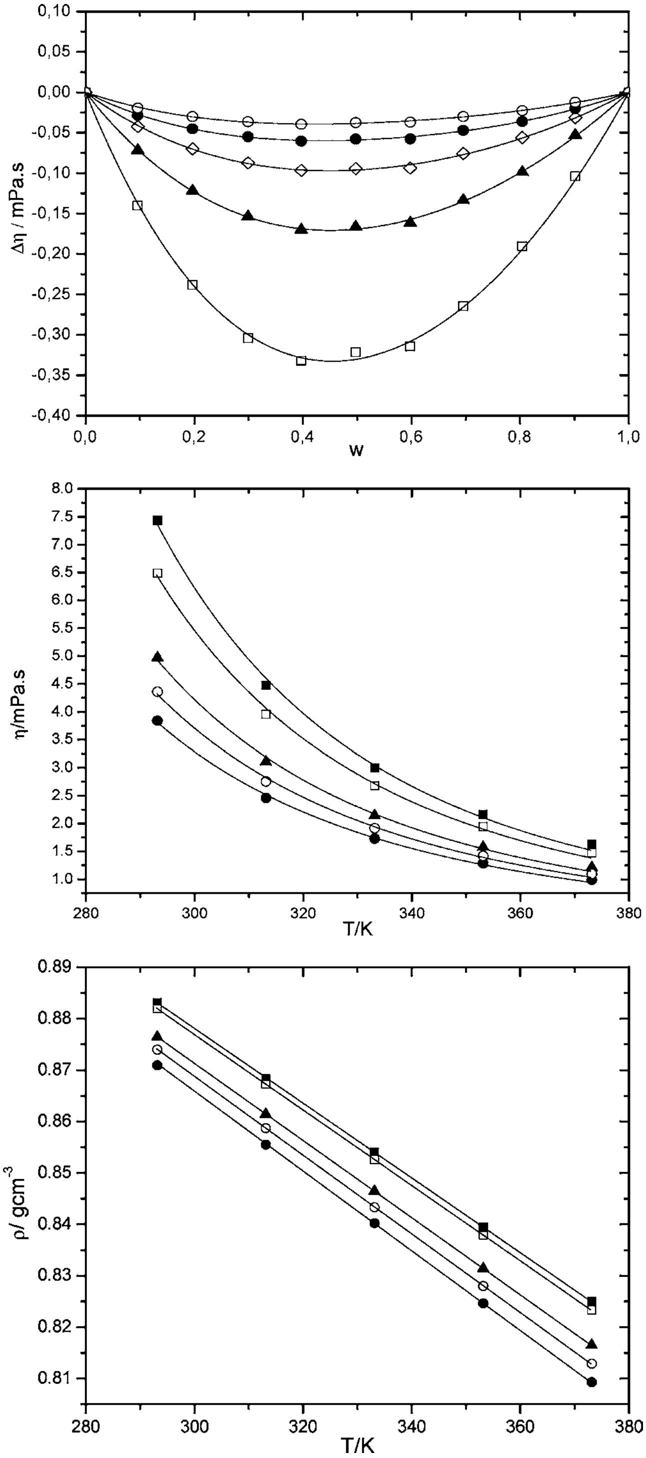

Figure 1. Top: viscosity deviation of the binary mixtures vs mass fraction of coconut biodiesel: □, at 293.15 K. ▲, at 313.15 K; ◇, at 333.15 K; ●, at 353.15 K; ○, at 373.15 K. Experimental results: w coconut biodiesel + (1 – w) colza biodiesel. Lines calculated from Redlich−Kister polynomials. Middle: temperature effect on dynamic viscosity for blends of coconut biodiesel + colza biodiesel: ■, pure colza biodiesel; □, blend at 100 w = 19.67 coconut biodiesel; ▲, blend at 100 w = 59.77 coconut biodiesel; ○, blend at 100 w = 80.39 coconut biodiesel; ●, pure coconut biodiesel. Temperature range T = (293.15 to 373.15) K. Data fitted to Andrade's type equation. Bottom: temperature effect on density for blends of coconut biodiesel + colza biodiesel: ■, pure colza biodiesel; ○, blend at 100 w = 19.67 coconut biodiesel; ▲, blend at 100 w = 59.77 coconut biodiesel; ○, blend at 100 w = 80.39 coconut biodiesel; ●, pure coconut biodiesel. Temperature range T = (293.15 to 373.15) K. Data fitted to a first-order polynomial equation.

the dependence on temperature of measured density and viscosity and also the dependency on composition of the calculated viscosity deviation of the coconut + colza system. It should be important to notice that a same behavior was observed for the coconut + soybean system.

As expected, the densities and viscosities decrease with increasing temperatures for all blends and pure compounds studied. It follows that coconut + colza and coconut + soybean biodiesel systems do not exhibit anomalous viscosity or density behavior, that is, show properties typical of the overwhelming majority of Newtonian liquids. It could be also observed that the viscosity values for the biodiesels studied in this work decrease in the following sequence: colza > soybean > coconut. This behavior is directly related to the composition of the biodiesels, for example, average carbon number, insaturation position, and glycerin content. These properties show a correlation with the viscosity properties of the biodiesels such as kinematic viscosity; see Tables 1 and 2. For densities, this sequence is the following: soybean ≈ colza > coconut, that is, density varies according to the molecular weight. Considering that the soybean and colza biodiesels mainly consisted of C18, that is slighter difference on the densities measured for these biodiesels. Nevertheless, coconut biodiesel presents in its composition smaller fatty acid methyl ester (C 12:0, C 14:0, and C 16:0); see Table 2.

The estimated parameters of the first-order polynomial equation (eq 6) are given in Table A (in the Supporting Information, SI), where A_0 and A_1 are constants specific for a temperature.

$$\rho = A_0 - A_1 T \qquad (6)$$

When the ratio of temperature to a liquid's critical temperature, T_r, is less than 0.7, kinematic viscosity is often represented by Andrade equation. A modified form of the Andrade equation, as proposed by Reid et al.,[13] was used, as it follows:

$$\ln(\eta) = A_0 + \frac{A_1}{\tau} \qquad (7)$$

Results of the regressions using eq 7 are shown in Table B (in the SI), where A_0 and A_1 are constants specific for a temperature.

Viscosity deviation values were correlated by means of the Redlich−Kister polynomial,[14] which for binary mixtures is

$$\Delta\eta = x_1(1 - x_1)\sum_{j=1}^{k} A_j(1 - 2x_1)^j \qquad (8)$$

where $\Delta\eta$ is the viscosity deviation, x_1 is the mole fraction, A_j is a parameter, and k is the degree of the polynomial expansion. A_j values were obtained by a nonlinear least-squares fitting procedure. The corresponding standard deviations are given by

$$\sigma(\Delta Y) = \sqrt{\frac{\sum \Delta Y_{\text{exp}} - \Delta Y_{\text{cal}}}{(n - p)}} \qquad (9)$$

where subscript cal refers to calculated data and n and p are the number of experimental points and number of parameters retained in the respective equation. The adjustable parameters, A_k, and standard deviation, σ, calculated using eq 9 for viscosity deviations are listed in Table C (in the SI).

The results shown in Figure 1 indicate that $\Delta\eta$ values are slightly negative for all temperatures and systems studied in this work and decrease with the increasing in temperature.

To determine the viscosity of the biodiesel using Ceriani's method we have used an ideal mixing rule, that is, a direct proportion between methyl esters contents and their pure

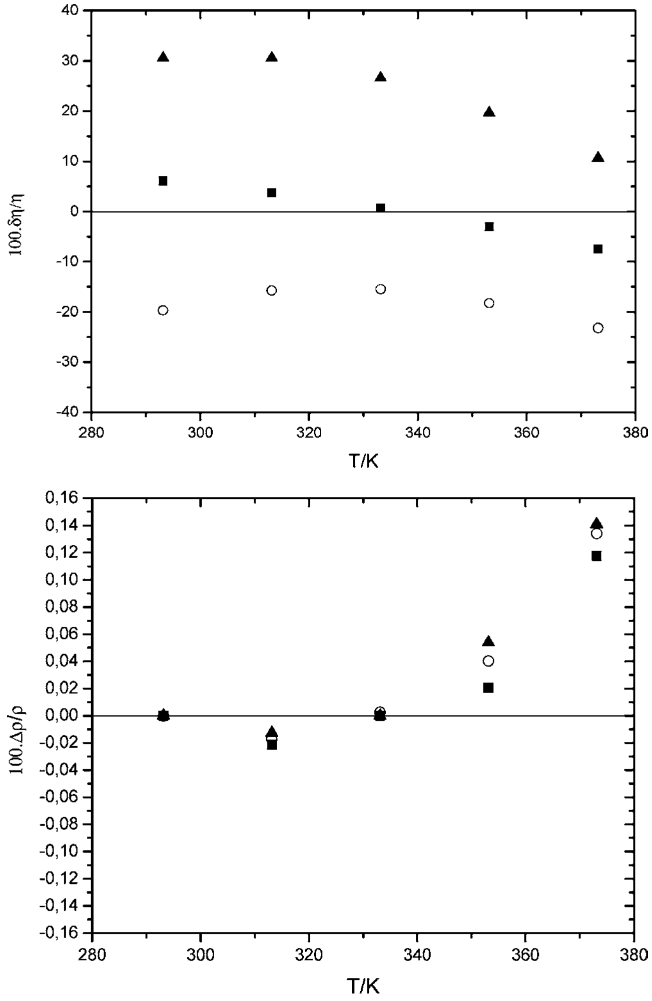

Figure 2. Top: deviation between experimental and predicted viscosity using Ceriani's method. ■, coconut biodiesel; ▲, colza biodiesel; and ○, soybean biodiesel. Bottom: prediction error for Racket equation. Properties were calculated for the Joback method. ■, coconut biodiesel; ▲, colza biodiesel; and ○, soybean biodiesel.

Table 6. Estimated Critical Properties of the Mixtures of Fatty Acids Methyl Esters, T_{cm}, P_{cm}, and V_{cm}, Obtained Using Lee−Kesler Mixing Rules

	T_{cm}	P_{cm}	V_{cm}
biodiesel	K	bar	$cm^3 \cdot mol^{-1}$
coconut	719.34	14.73	836.99
soybean	759.07	13.11	1024.96
colza	759.13	12.88	1040.36

viscosity (see Table 2). Table D (in the SI) shows the constants' values used on the prediction of viscosity of fatty compounds. Figure 2 shows the comparison between our viscosity measurements and the values obtained by using Ceriani's method for pure biodiesels studied on this work. It could be observed that there is a maximum relative deviation of 30 % for colza and soybean biodiesel. The best agreement was obtained for coconut biodiesel with a maximum relative deviation less than 10 %.

Table 6 shows some physicochemical properties for the mixtures studied on this work. The critical parameters of the fatty acid methyl esters obtained using Joback's method are given in Table E (in Supporting Information). In Table 7 the values for Rackett compressibility factor (Z_{RA}) can be observed. Figure 2 shows the correlation between Joback's method and our experimental data. It could be observed a good agreement, with a maximum relative deviation in the order to 0.14 %.

Table 7. Rackett Compressibility Factor Z_{RA} for Methyl Esters Obtained Using the Spencer and Danner Method

biodiesel	Rackett compressibility factor Z_{RA}
coconut	0.223
soybean	0.219
colza	0.220

Conclusions

In this paper, the thermophysical property data (density and viscosity) of binary mixtures of coconut + colza and coconut + soybean biodiesels were measured at $T = (293.15, 313.15, 333.15, 353.15, \text{and } 373.15)$ K. Densities and viscosities data were correlated with *R*-square adjustment and Andrade equation, respectively, with good agreement. The deviation viscosities ($\Delta\eta$) have been calculated from the experimental data. Viscosity deviation was calculated using experimental data. All systems measured exhibited negative viscosity deviation values, which decrease with rise in temperature. The deviations have been correlated using the Redlich−Kister polynomial equation. We have used two estimative approaches to predict densities and viscosities of the systems studied in this work. Joback's method for predicting density (maximum deviation 0.14 %) and a group contribution model proposed by Ceriani et al., with a maximum deviation of 30 %.

Supporting Information Available:

Estimated parameters for density, viscosity, and viscosity deviation of the binary mixtures (coconut biodiesel + Soybean's biodiesel and coconut biodiesel + colza biodiesel) are shown in Tables A to C. Also, the constants of group contribution model for prediction of viscosity of fatty compounds and their critical properties (Tables D and E). This material is available free of charge via the Internet at http://pubs.acs.org.

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