

Volumetric Properties of Pseudo-Binary and Ternary Mixtures with Biodiesel

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To better understand the behavior of pseudo-ternary biodiesel blends, the study of pseudo-binary and ternary mixtures with biodiesel, diesel fuel and benzene with components perfectly miscible on the whole composition range was considered to be of interest. The densities of ternary biodiesel + diesel fuel + benzene mixtures under atmospheric pressure, covering the whole composition range and for temperature ranging from 293.15 K to 323.15 K were reported. The accuracy of some mixing rules to predict and correlate the density of ternary biodiesel + diesel fuel + benzene mixture was tested. In order to analyze the blending process, the ternary excess molar volume, V_{123}^E , was derived from experimental density data of the ternary mixtures and pure components. Cibulka equation was used to correlate the ternary V_{123}^E data using the Redlich–Kister binary parameters.

Keywords: biodiesel, diesel fuel, density, excess molar volume, ternary mixture.

The interest regarding biofuels, especially biodiesel, as a way to reduce environment pollution, significantly increased in the last years [1- 6].

Binary and ternary blends of biodiesel and diesel fuel are of interest to support the efforts to diversify the biofuels types used in transportation. A relatively small number of works on the properties of ternary blends with biodiesel [7 - 12] were presented in literature.

To better understand the behavior of ternary blends with biodiesel, the study of pseudo-ternary biodiesel + diesel fuel + benzene mixture with components perfectly miscible on the whole composition range was considered to be of interest. The knowledge of the physico-chemical properties is essential in the development of a fuel with properties required by the standards.

Density is one of the most studied properties of biodiesel with diesel fuel mixtures [13 -15], because of its impact on fuel quality. Since density influences the processes taking place in the internal combustion engine, and also production, transportation, and distribution processes, experimental data and correlations for this property, together with the estimation of derived volumetric properties will be of interest.

In the present paper we report densities of pseudo-ternary biodiesel + diesel fuel + benzene mixtures and corresponding pseudo-binaries mixtures under atmospheric pressure, covering the whole composition

range and for temperature ranging from 293.15 K to 323.15 K. The aim of this study was to evaluate the accuracy of some mixing rules to predict and correlate the density of pseudo-ternary biodiesel + diesel fuel + benzene mixture, depending on its composition or temperature. Equations for biodiesel mixtures properties calculation were extended from binary to ternary mixtures. In order to analyze the biodiesel + diesel fuel + benzene blending process, the ternary excess molar volume, V_{123}^E , was derived from experimental density data of the ternary mixtures and pure components. Excess molar volumes of the corresponding binary mixtures were fitted to the Redlich-Kister polynomial equations. Cibulka equation was used to correlate the ternary V_{123}^E data using the Redlich–Kister binary parameters.

Experimental part

Biodiesel was obtained by transesterification from rapeseed oil and methanol according to a previously presented procedure [16]. Diesel fuel was provided from/ by a local fuel supplier and complies with European standards; benzene of 99.7 % purity was purchased from Merck. Physicochemical properties of the ternary system components, biodiesel, diesel fuel and benzene are presented in table 1, compared to literature data, generally the agreement being good.

Table 1

DENSITY (ρ), REFRACTIVE INDEX (n_D), MOLAR WEIGHT (M), KYNEMATIC VISCOSITY (ν), AND FLASH POINT VALUES (FP) OF THE PSEUDO PURE AND PURE COMPONENTS, AND THEIR COMPARISON WITH LITERATURE

Component	ρ at 20°C (g/cm ³)			n_D at 25°C			M (g/mol)		
	exp	lit	reference	exp	lit	reference	exp	lit	reference
Biodiesel	0.8823	0.8830	[17]	1.4540	1.4541	[19]	294.32	294.80	[22]
Diesel fuel	0.8414	0.8424	[15]	1.4650	1.4830	[20]	213.89	211.00	[23]
Benzene	0.8772	0.8789	[18]	1.4968	1.4969	[21]		78.11	
	η at 40°C (mm ² /s)			FP (°C)			ester content (%)		
	exp	lit	reference	exp	lit	reference	exp	lit	reference
Biodiesel	4.6439	4.5	[24]	131	139	[15]	96.80	97.10	[15]
Diesel fuel	3.5439	3.40	[24]	60	58	[15]	-	-	-
Benzene	0.5820	0.5762	[25]		-11	[26]	-	-	-

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Table 2
FATTY ACID METHYL ESTER PROFILE OF BIODIESEL

fatty acid methyl ester*	16:0	16:1	18:0	18:1	18:2	18:3	20:0	20:5	22:1
% w/w	4.91	0.27	1.56	64.27	20.38	6.97	1.17	0.47	0.30

*The first digit indicates the number of carbon atoms in the hydrocarbonated chain of the ester, the second digit indicates the number of double

bonds from the hydrocarbonated chain.

The mean molar weight of biodiesel was determined based on its fatty acid methyl ester concentration profile determined by gas chromatography (table 2) (Clarus 500 GC chromatograph equipped with a FID detector and capillary column SGE BPX70 of high polarity, with polysiloxane as stationary phase; hydrogen flow rate 20 mL/min). The mean molar weight of diesel fuel was determined using the cryoscopic method with benzene as solvent, as presented elsewhere [16]. The refrigeration of pure benzene and of diesel fuel+benzene blends was realized on an ice bath. The accuracy of the thermometer was ± 0.3 °C. Three measurements of the melting point of diesel fuel+benzene solutions were made. The mean value was taking into account for diesel fuel molar weight calculation.

Ternary biodiesel+diesel fuel+benzene and corresponding binary mixtures were prepared at room temperature by weighting (OHAUS analytical balance with a precision of ± 0.0001 g). The mixtures were prepared to cover the entire composition range. All mixtures were completely miscible. In total 35 samples (32 mixtures) were used for density measurements of ternary system. In order to prevent evaporation, the mixtures were prepared into stoppered bottles.

The experimental measurements of density were carried out using an Anton Paar SVM 3000 density meter at 293.15, 298.15, 303.15, 308.15, 313.15 K, 318.15 and 323.15K. This equipment contains an U-shaped vibrating tube measurement cell to determine the density. It had an integrated Peltier thermostat, whose accuracy of temperature is ± 0.02 °C. The uncertainty for density measurement is ± 0.0005 g/cm³. Doubled distilled and degassed water and dry air were used for the calibration of the density meter. The apparatus was calibrated before each series of measurements. All measurements were repeated three times, and the results were averaged.

Several mixing rules to predict and correlate the density of ternary mixture biodiesel+diesel fuel+benzene, depending on its composition or temperature were tested. Equations used to predict the density of binary mixture with biodiesel were extended for ternary mixtures:

-Kay mixing rule:

$$\rho = x_1\rho_1 + x_2\rho_2 + x_3\rho_3 \quad (1)$$

where ρ is the density of the mixture (g/cm³), ρ_1 , ρ_2 and ρ_3 are the densities of components 1, 2 and 3, respectively (g/cm³), x_1 , x_2 and x_3 are the molar fractions of the three components of the mixture.

-equation for petroleum fractions [27]:

$$\rho = \frac{x_1M_1 + x_2M_2 + x_3M_3}{\frac{x_1M_1}{\rho_1} + \frac{x_2M_2}{\rho_2} + \frac{x_3M_3}{\rho_3}} \quad (2)$$

where M_1 , M_2 and M_3 are molar weights (g/mol) of the three components of the mixture.

-empirical equation used to correlate the density of binary biodiesel+diesel fuel mixture with temperature:

$$\rho = aT + b \quad (3)$$

where T is temperature (K), a (g/cm³K) and b (g/cm³) are adjustable parameters.

The excess molar volumes were obtained from experimental density data of the mixtures, and of pure components, using the following equation:

$$V^E = \sum_{i=1}^n \frac{x_i M_i}{\rho} - \sum_{i=1}^n \frac{x_i M_i}{\rho_i} \quad (4)$$

where V^E is the experimental excess molar volume of the mixture (cm³/mol), ρ is the density of the ternary mixture, x_i , M_i and ρ_i are the molar fraction, molar weight and density of the i component, n is the number of the components in the mixture.

Cibulka equation was used to correlate the ternary excess molar volume, V_{123}^E , using the Redlich-Kister binary parameters:

$$V_{123}^E = V_{bin}^E + x_i x_j (1 - x_i - x_j) (B_0 + B_1 x_i + B_2 x_j) \quad (5)$$

where V_{123}^E represents the ternary excess molar volume, V_{bin}^E represents the binary contribution to the excess ternary molar volume; B_0 , B_1 , and B_2 represent adjustable parameters; the last term of eq.(5) represents the ternary contribution to the ternary excess molar volume.

The binary contribution to the excess ternary molar volume, V_{bin}^E , was calculated with the equation [28]:

$$V_{bin}^E = \sum_{i,j} V_{ij}^E(x_i, x_j) \quad (6)$$

where $V_{ij}^E(x_i, x_j)$ is the value of the excess molar volume of the corresponding binary mixtures fitted to the Redlich-Kister polynomial equation, in which the molar fractions are those of the ternary mixtures. Redlich-Kister polynomial equation is:

$$V_{ij}^E = x_i x_j \sum_{k=0}^4 A_k (x_i - x_j)^{k-1} \quad (7)$$

where A_k denotes adjustable parameters, k is the degree of polynomial. The set of A_k parameters for each binary mixture (biodiesel+diesel fuel, biodiesel+benzene, diesel fuel+benzene) was calculated by fitting the experimental excess binary molar volumes in eq. (7).

The accuracy of the models used to calculate the density of the ternary system was evaluated using the average relative deviation, ARD (%):

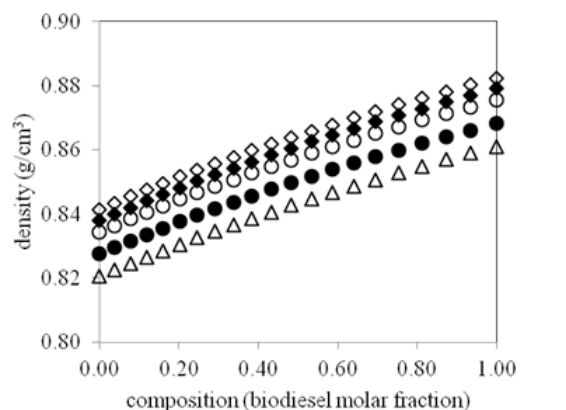
$$ARD = \frac{100}{N} \sum_{i=1}^N \frac{|\rho_{exp,i} - \rho_{cal,i}|}{\rho_{exp,i}} \quad (8)$$

where ρ_{exp} is the experimental value of the density, ρ_{cal} is the calculated value of the density, N represents the number of experimental points.

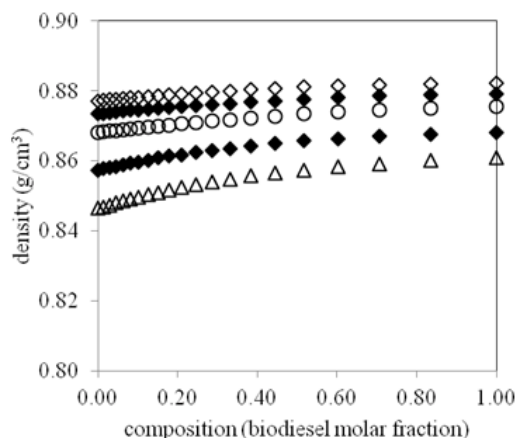
Standard deviation, SD was used in order to determine the Redlich-Kister and Cibulka equations parameters:

$$SD = \sqrt{\frac{\sum_{i=1}^N (V_{cal,i}^E - V_{exp,i}^E)^2}{N - m}} \quad (9)$$

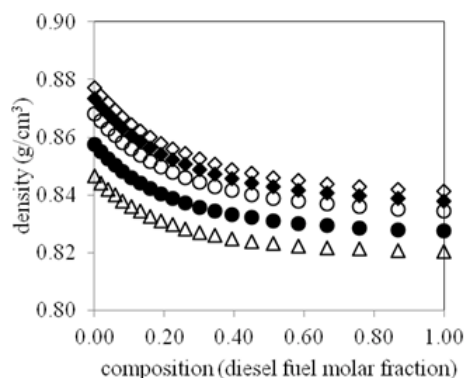
where V_{cal}^E and V_{exp}^E represent the calculated and respectively, the experimental value of the excess molar volume, m is the number of adjustable parameters of eqs. (5) and (7), respectively.



a) biodiesel + diesel fuel



b) biodiesel + benzene



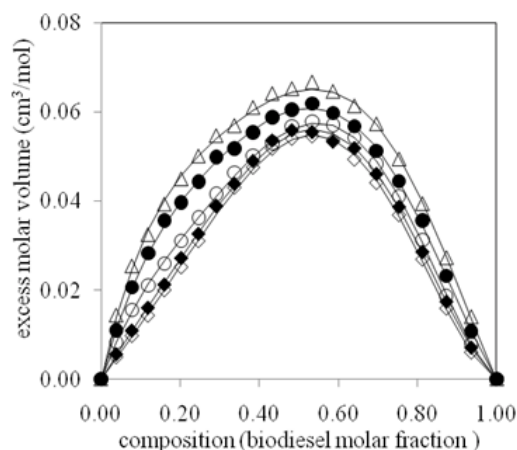
c) biodiesel fuel+ benzene

Fig. 1. Density versus composition for biodiesel+diesel fuel (a), biodiesel+benzene (b), and diesel fuel +benzene (c) binary systems: $T = 293.15\text{ K}$, $\diamond$; 298.15 K , $\blacklozenge$; 303.15 K , $\circ$; 313.15 K , $\bullet$; 323.15 K , Δ

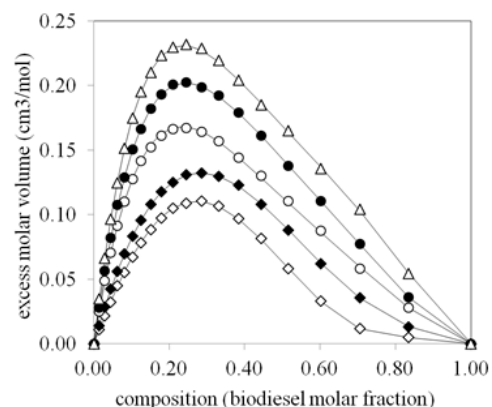
Results and discussions

Experimental results of the density and excess molar volume V^E_{12} for the binary systems biodiesel+diesel fuel, biodiesel+benzene, and diesel fuel+benzene, over the entire composition range and for temperature ranging from 293.15 K to 323.15 K are presented in figure 1 and 2, respectively.

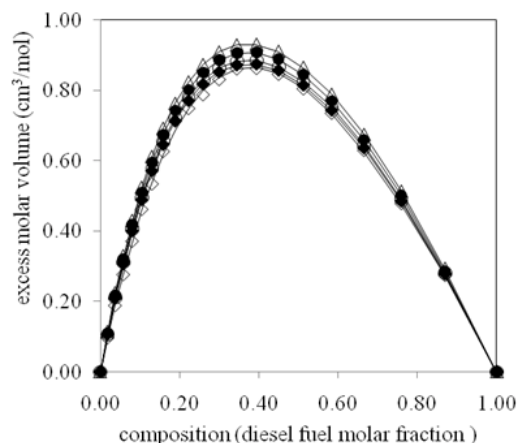
As shown in figure 1, a monotonously variation of density with composition can be observed for the binary investigated systems. Density decreases with temperature increasing. The excess molar volumes corresponding to pseudo-binary biodiesel+diesel fuel, biodiesel+benzene and diesel fuel+benzene mixtures are positive over the whole composition and temperature range (fig. 2). This would indicate the presence of weak dispersion type interactions. Positive excess molar volume was reported



b) biodiesel + benzene



b) biodiesel + benzene



c) biodiesel fuel+ benzene

Fig. 2. Excess molar volume, V^E_{12} against composition for pseudo-binary mixtures biodiesel+diesel fuel (a), biodiesel+benzene (b) and diesel fuel+benzene (c): $T = 293.15\text{ K}$, $\diamond$; 298.15 K , $\blacklozenge$; 303.15 K , $\circ$; 313.15 K , $\bullet$; 323.15 K , Δ . Solid lines correspond to the fit using equation (7)

in literature for the pseudo-binary system biodiesel + benzene [29].

Table 3 presents density and excess molar volume V^E_{123} for the ternary biodiesel + diesel fuel + benzene system. As an example, the density variation with composition for the ternary biodiesel + diesel fuel + benzene mixture at 298.15 K is presented in figure 3. As seen in table 3 and figure 3, the density of the ternary mixture varies monotonously, without extreme points, as in the case of the corresponding binary systems, with values in the range limited by the densities of the pure components. The same behavior was registered for all investigated temperatures.

Table 3

EXPERIMENTAL DENSITY VALUES (g/cm^3) AND V^E (cm^3/mol) OF TERNARY BIODIESEL (1) + DIESEL FUEL (2) + BENZENE (3) MIXTURES FOR TEMPERATURE RANGING FROM 293.15 K TO 323.15 K

Composition	Temperature (K)											
	293.15 K			298.15 K			303.15 K			308.15 K		
x_1	x_2	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E	ρ	V^E	ρ
0.6128	0.1005	0.8737	1.1703	0.8708	1.0661	0.8658	1.4455	0.8588	2.4155	0.8513	2.4146	0.8476
0.4387	0.2877	0.8607	2.4998	0.8575	2.4777	0.8528	2.7790	0.8494	2.6924	0.8456	2.7308	0.8382
0.2053	0.5385	0.8477	2.5718	0.8445	2.5285	0.8401	2.7426	0.8358	2.9386	0.8321	2.9596	0.8247
0.0656	0.6887	0.8435	1.4455	0.8403	1.3807	0.8327	2.4370	0.8270	3.0207	0.8233	3.0488	0.8196
0.0566	0.5196	0.8445	1.8215	0.8414	1.7432	0.8338	2.6206	0.8300	2.6397	0.8261	2.6884	0.8244
0.0498	0.3915	0.8466	1.8871	0.8440	1.7170	0.8404	1.6451	0.8336	2.2275	0.8296	2.2413	0.8276
0.1011	0.3314	0.8498	2.0863	0.8454	2.2855	0.8433	2.1127	0.8383	2.1226	0.8343	2.1258	0.8322
0.2390	0.3135	0.8525	2.9042	0.8516	2.3260	0.8477	2.3481	0.8438	2.3660	0.8400	2.3591	0.8322
0.0401	0.2102	0.8557	1.2004	0.8518	1.2627	0.8477	1.1959	0.8416	1.5141	0.8372	1.5293	0.8355
0.2087	0.2033	0.8575	2.2326	0.8529	2.4906	0.8505	2.1635	0.8478	1.8863	0.8439	1.8602	0.8397
0.1369	0.1796	0.8588	1.6291	0.8536	1.9514	0.8504	1.7798	0.8472	1.5979	0.8430	1.5979	0.8388
0.0363	0.1436	0.8601	0.9721	0.8566	0.9673	0.8524	0.9225	0.8461	1.1798	0.8415	1.1865	0.8370
0.2652	0.1391	0.8638	1.8005	0.8594	2.0208	0.8562	1.8528	0.8526	1.7542	0.8486	1.7451	0.8445
0.0738	0.0968	0.8652	0.8339	0.8609	0.9477	0.8557	1.0465	0.8511	1.0498	0.8466	1.0378	0.8421
0.4431	0.0830	0.8717	1.3301	0.8684	1.3324	0.8619	1.9830	0.8580	1.9735	0.8541	1.9782	0.8502
0.1665	0.0546	0.8706	0.8068	0.8667	0.8810	0.8626	0.8345	0.8566	1.0878	0.8522	1.0837	0.8478
0.0310	0.0407	0.8716	0.3000	0.8676	0.3469	0.8625	0.3696	0.8562	0.3306	0.8513	0.3209	0.8463
0.5237	0.1962	0.8668	1.9613	0.8638	1.8848	0.8584	2.3883	0.8541	2.5561	0.8504	2.5601	0.8466
0.1340	0.6152	0.8452	2.1049	0.8419	2.0784	0.8378	2.2062	0.8311	3.0627	0.8274	3.0880	0.8236
0.3574	0.3750	0.8559	2.6555	0.8524	2.7121	0.8486	2.7563	0.8448	2.7959	0.8411	2.8090	0.8373
0.2797	0.4585	0.8512	2.7790	0.8478	2.8000	0.8440	2.8477	0.8402	2.8965	0.8365	2.9139	0.8327
0.1152	0.4534	0.8462	2.3935	0.8425	2.4636	0.8387	2.4691	0.8349	2.4794	0.8312	2.4561	0.8274
0.1760	0.3847	0.8490	2.7267	0.8472	2.3602	0.8434	2.3618	0.8396	2.3635	0.8359	2.3536	0.8321
0.0900	0.2361	0.8538	1.7754	0.8500	1.8400	0.8463	1.7621	0.8435	1.5150	0.8398	1.4248	0.8360
0.1232	0.1078	0.8641	1.1845	0.8599	1.3052	0.8565	1.1515	0.8534	0.9360	0.8497	0.8169	0.8459
0.0678	0.0444	0.8710	0.4722	0.8672	0.5049	0.8630	0.4338	0.8592	0.2969	0.8555	0.1447	0.8517
0.3236	0.0707	0.8709	1.1733	0.8669	1.3330	0.8632	1.2608	0.8582	1.4481	0.8544	1.3899	0.8505
0.0335	0.0879	0.8659	0.6003	0.8619	0.6563	0.8577	0.5892	0.8539	0.4593	0.8500	0.3385	0.8462
0.3724	0.1628	0.8642	2.1327	0.8603	2.2804	0.8574	2.0502	0.8535	2.0496	0.8497	2.0331	0.8457
0.2349	0.0616	0.8708	0.9664	0.8668	1.0779	0.8620	1.1863	0.8582	1.0955	0.8542	1.0474	0.8504
0.1121	0.0490	0.8707	0.5936	0.8667	0.6809	0.8624	0.6722	0.8584	0.5774	0.8547	0.4385	0.8507
0.0444	0.2910	0.8515	1.4443	0.8475	1.5359	0.8440	1.4228	0.8386	1.6507	0.8349	1.5651	0.8311

Figure 4 highlights the influence of temperature on the density of the ternary mixture. It can be observed that the density of the pseudo-ternary biodiesel+diesel fuel+benzene mixtures decreases with temperature increasing, as in the case of the corresponding pseudo-binary systems. The same behavior was registered for the density of biodiesel+diesel fuel system [13, 15].

The excess molar volume of the pseudo-ternary biodiesel+diesel fuel+benzene system is positive over the whole composition and temperature range (table 3). The values of the excess molar volumes become more positive as temperature increases.

Density calculation

Eqs. (1) and (2) were tested in order to estimate the density of ternary system biodiesel+diesel fuel+benzene at fixed temperatures. The errors in density estimation with eq.(1) and eq.(2), respectively, are positive, meaning the

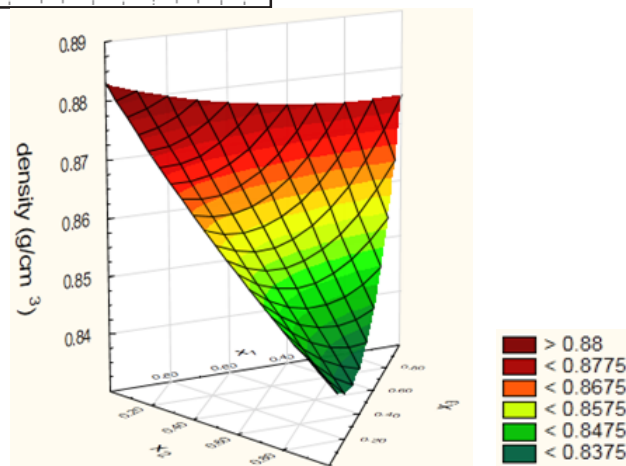


Fig. 3. Density versus composition for biodiesel (1) + diesel fuel (2) + benzene (3) ternary system at 298.15 K

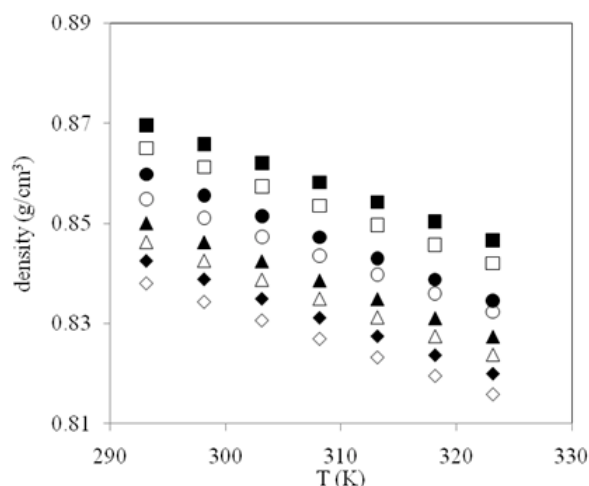


Fig. 4. Density vs temperature for biodiesel(1)+diesel fuel(2)+benzene(3) ternary system at different composition; ■: $x_1=0.2349$, $x_2=0.0616$; □: $x_1=0.3724$, $x_2=0.1628$; ●: $x_1=0.1369$, $x_2=0.1796$; ○: $x_1=0.0900$, $x_2=0.2361$; ▲: $x_1=0.0444$, $x_2=0.2910$; △: $x_1=0.1152$, $x_2=0.4534$; ◆: $x_1=0.1340$, $x_2=0.6152$; ◇: $x_1=0.0656$, $x_2=0.6887$

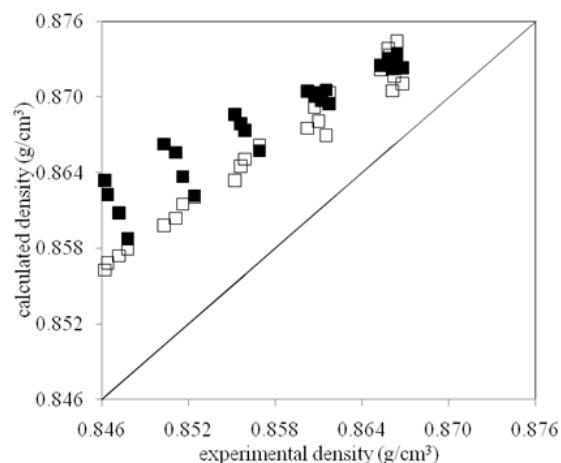


Fig. 5. Calculated vs experimental density at 298.15 K for ternary system biodiesel+diesel fuel+benzene; ■eq. (1); □ eq. (2)

Parameter	Temperature (K)				
	293.15	298.15	303.15	313.15	323.15
Biodiesel+diesel fuel					
A_0	0.2173	0.2223	0.2274	0.2429	0.2595
A_1	0.0513	0.0534	0.0619	0.0302	0.0350
A_2	-0.1342	-0.1183	-0.0729	-0.0098	0.0402
A_3	-0.0927	-0.0953	-0.1468	-0.1238	-0.1429
SD	0.0007	0.0007	0.0004	0.0006	0.0008
Biodiesel+benzene					
A_0	0.2534	0.3713	0.4526	0.5725	0.6744
A_1	-0.6313	-0.5858	-0.5419	-0.6393	-0.6185
A_2	0.2325	0.1908	0.5003	0.5371	0.6994
A_3	0.3335	0.1041	-0.3771	-0.4136	-0.5911
SD	0.0005	0.0003	0.0011	0.0006	0.0009
Diesel fuel+benzene					
A_0	3.2661	3.2898	3.3309	3.4131	3.4938
A_1	-1.5691	-1.5122	-1.5355	-1.5836	-1.6316
A_2	0.5851	0.8270	0.8438	0.8793	0.9155
A_3	-0.0268	-0.4574	-0.4687	-0.4930	-0.5179
SD	0.0079	0.0017	0.0018	0.0019	0.0020
Biodiesel+diesel fuel+benzene					
B_0	74.57	87.51	63.35	-22.80	-177.13
B_1	-37.14	-54.62	-0.13	151.27	499.54
B_2	16.19	-12.25	26.75	170.88	354.84
SD	0.0735	0.14339	0.3164	0.5680	0.9447

Table 4
VALUES OF CIBULKA (EQ.5) AND REDLICH-KISTER PARAMETERS (EQ.7) AND STANDARD DEVIATION, SD (cm³/mol), FOR V^E AT DIFFERENT TEMPERATURES

estimated density values are slightly greater than the experimental ones. The accuracy of eq.(2) is better than that of eq.(1) at all investigated temperatures, the maximum ARD registered with eq.(1) was 1.2030%, and with eq.(2) was 0.9857%. Better accuracy of eq.(2) with respect to eq.(1) results from figure 5 where calculated versus experimental values of density are represented at 298.15 K. The same behaviour was registered at all investigated temperatures.

The ARD corresponding to density calculation at different temperatures using eq. (3) varied between 0.2104 and 2.0945%, indicating a relatively good accuracy of the linear relationship between density and temperature for this ternary system.

Excess molar volume calculation

Excess molar volumes of the ternary system biodiesel+diesel fuel+benzene were calculated from the experimental densities of the mixtures, the density and the molar weight of the components of the system. The experimental values of the excess molar volume, V_{123}^E of the ternary mixture biodiesel+diesel fuel+benzene

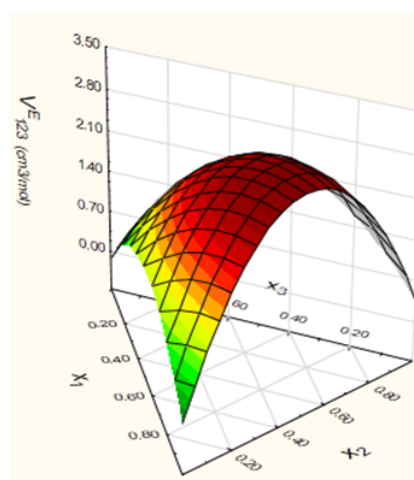


Fig.6. Excess molar volume, V_{123}^E for pseudo-ternary mixture biodiesel+diesel fuel+benzene at 298.15 K.

calculated with eq. (4) are positive (table 3) over the entire composition range and at all investigated temperatures, suggesting no packing effect; they present deviations of max 2% from ideal mixture volume. Ternary excess molar volumes, V_{123}^E , were fitted to Cibulka equation (eq. 5), taking

into account the binary, and the ternary contribution, respectively. Values of the adjustable parameters, A_k calculated by fitting the experimental excess binary molar volumes V_{ij}^E , in eq.(7), and respectively the adjustable parameters B_0, B_1, B_2 calculated by fitting the experimental excess ternary molar volumes, V_{123}^E , in eq.(5) are reported in table 4, along with the corresponding standard deviations. As an example, figure 6 shows a graphical representation of calculated V_{123}^E on the whole composition domain at 298.15 K. It can be observed the quite regular shape of the V_{123}^E surface.

Conclusions

Experimental results of the density and excess molar volume for the pseudo-ternary mixture biodiesel + diesel fuel + benzene and corresponding binary mixtures over the entire composition range and for temperature ranging from 293.15 K to 323.15 K were reported. The density of the pseudo-ternary mixture varies monotonously, without extreme points, as in the case of the corresponding binary systems, with values in the range limited by the densities of the pure components. The density of each ternary biodiesel + diesel fuel + benzene mixture decreases with temperature increasing. The density of the pseudo-ternary system can be accurately predicted in the investigated temperature range (ARD less than 0.9857%) with a mixing equation, based on the densities and molar weights of mixture components. The accuracy of Kay mixing rule is slightly lower (ARD less than 1.2030%). The density can be accurately calculated (ARD less than 2.09%) at different temperatures using a linear relationship between density and temperature for this pseudo-ternary system. The excess molar volume, V_{123}^E of the pseudo-ternary system biodiesel + diesel fuel + benzene is positive over the entire composition range and at all investigated temperatures showing deviations of max 2% from ideal mixture volume.

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