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REFRACTIVE INDICES FOR BIODIESEL MIXTURES

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Au fost determinați experimental indicii de refracție pentru amestecuri biodiesel + motorină şi biodiesel + toluen, realizate cu trei tipuri de biodiesel la temperaturile de 298,15K; 303,15K; 313,15K; şi 323,15K, pe întreg domeniul de compoziție. Datele au fost utilizate pentru a obține ecuații de calcul ale unor proprietăți fizico-chimice importante ale amestecurilor, cum ar fi densitatea şi viscozitatea, mai dificil de obținut experimental. De asemenea, s-a testat capacitatea de predicție a indicelui de refracție cu ecuațiile tradiționale Lorentz-Lorenz, Dale-Gladstone, Eykman, Newton şi Kay, aplicate în acest caz amestecurilor cu biocombustibil.

Refractive indices for biodiesel + diesel fuel and biodiesel + toluene mixtures using three different types of biodiesel at 298.15K, 303.15K, 313.15K and 323.15K on the entire range of composition were determined experimentally. The data have been used to obtain the equations to calculate some important physical properties of mixtures like density or viscosity, more difficult to obtain experimentally. Also, the predictive computing capacity of refractive index applied to biofuel mixtures was tested with the traditional equations Lorentz-Lorenz, Dale-Gladstone, Eykman, Newton and Kay.

Keywords: refractive index, correlative equations, biodiesel, biofuel

1. Introduction

Biofuels are becoming increasingly important. Biodiesel, derived from vegetable oils or animal fats is a promising alternative to conventional diesel fuel. Biodiesel is obtained by the transesterification process, which is a chemical reaction between triglycerides (oil) and a monoalcohol in the presence of a catalyst to produce monoesters (biodiesel) and glycerin as by-product [1].

Biodiesel can be blended with diesel fuel and used in diesel engines with little or no modifications. There are several key properties which need to be known before using biodiesel-diesel fuel mixtures in a diesel engine. These

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properties include viscosity, density, pour point, flash point and range of distillation temperatures. Kinematic viscosity is one of the most important fuel properties. The use of biodiesel in diesel engines is limited mainly by the viscosity [2]. Fuel droplet size affects the quality of combustion [3, 4, 5]. Low viscosity can cause leaks in the fuel system, while high viscosity leads to incomplete combustion, increased deposits in the engine and the need of more power to fuel pump [2, 6, 7].

Using biodiesel has advantages: helps to reduce pollution emissions in the atmosphere, is renewable in nature and safer to handle, has no aromatic compounds, practically is free of sulfur. Oxygen atoms from the molecule of biodiesel lead to a better combustion [8].

However, biodiesel has several problems when compared with diesel fuel: higher emissions of particulate matters and nitrogen oxides [9,10] and higher production costs [11]. Last problem related to costs could be partially solved by using cooking oil as raw material in the process of transesterification and by valorization of by-products (glycerol).

It is estimated that biodiesel/bioethanol could replace 10% of diesel fuel demand in Europe and 5% of the demand for Southeast Asian gasoline [12].

In a previous paper we have presented the density and viscosity properties of biodiesel blends [13]. The purpose of the present study is to present refractive index experimental data for binary mixtures of biodiesel with diesel fuel or toluene and to correlate them both with composition and temperature as with various properties. In the case of biodiesel and its mixtures, refractive index could be used to predict some important physical properties such as density or viscosity. The experimental data were utilized to test the applicability of traditionally predictive equations of refractive index to systems with biodiesel. [14].

To the best of our knowledge, only few data were presented in literature regarding the refractive index of biodiesel and its mixtures [15].

2. Experimental

Chemicals

In this study, three samples of rapeseed oil were used in the production of methyl esters (biodiesel). Biodiesel was synthesized in the laboratory by transesterification method using methanol (CH_3OH) as an alcohol and potassium hydroxide (KOH) as catalyst [16]. The molar ratio methanol/oil was 6:1 and the catalyst was 1% by weight based on oil. Alcohol and catalyst mixture was added to the flask which already had 200 grams of oil. Installation is equipped with a flat-bottomed flask with water cooled condenser and a stirrer. Magnetic stirring took place for 2 hours at a temperature of 60 ^{o}C, stirring vigorously. The reaction mixture was transferred later in a separating funnel and after 12 hours was separated in two phases: biodiesel and glycerol. Biodiesel was repeatedly washed with distilled water at 100 ^{o}C until the pH of washing water was the same as distilled water. To remove residual water, biodiesel was dried over anhydrous sodium sulphate and finally filtered under reduced pressure to obtain the final product. Toluene (99.7%) was supplied by Merck. Methanol and potassium hydroxide were provided by Lachner. Commercially available diesel fuel was obtained from a local company. Vegetable oils were obtained locally. Fatty acid components of methyl esters were identified using gas- chromatography [17].

The characteristic properties of the three types of biodiesel utilized in this work, for diesel fuel and toluene are presented in Table 1.

Apparatus and procedure

Three types of biodiesel made from rapeseed oil, noted in text as Bio_I, Bio_{II}, Bio_{III}, have been used. The mixtures of biodiesel with diesel fuel or toluene were prepared by volume fractions of 0.05, 0.10, 0.15, 0.20, 0.25, 0.30, 0.35, 0.40, 0.45, 0.50, 0.55, 0.60, 0.65, 0.70, 0.75, 0.80, 0.85, 0.90 and 0.95, the usual procedure for fuel mixtures [13,18,19].

The mixtures of biodiesel are in fact pseudo-binary mixture because the biodiesel is itself a mixture of esters and not a pure compound [13,20]. The diesel fuel represents a mixture of saturated hydrocarbons with small quantities of aromatic hydrocarbons, too.

All mixtures were completely miscible. Each experiment was repeated three times to evaluate experimental errors.

Refractive index of the mixtures was measured with a refractometer Abbé, Atago 3T type. The reproducibility of refractive index data was of 10^{-4} and the thermostated bath had an accuracy of ± 0.05 °C.

Table 1

Properties of biodiesel, diesel fuel and toluene

Property	Units	Bio_I	Bio_{II}	Bio_{III}	Diesel fuel	Toluene
Density at 20°C	kg/m^3	0.8840	0.8806	0.8823	0.8414	0.8667
Kinematic viscosity at 40°C	mm^2/s	4.6715	5.0111	4.6439	3.5439	0.6860
Water content	mg/kg	258.36	230	200	130	-
Sulfur content	mg/kg	0.00	0.00	0.00	0.14	-
Flash point	°C	140	148	130	60	-
Methylic esters of fatty acids	% mass	96.50	97.90	97.00	3.0	-

3. Results and discussions

The refractive index experimental values of the of pseudo-binary mixtures of biodiesel + diesel fuel, biodiesel + toluene with three different types of biodiesel at 298.15, 303.15, 313.15K and 323.15K are presented in Table 2.

The experimental data was utilized to verify the predictive capacity of different proposed equations:

Lorentz-Lorenz:

$$\frac{n_{Dm}^2 - 1}{n_{Dm}^2 + 2} = \sum_{i=1}^{n}\left[v_i\left(\frac{n_{Di}^2 - 1}{n_{Di}^2 + 2}\right)\right] \tag{1}$$

Dale-Gladstone:

$$n_{Dm} - 1 = \sum_{i=1}^{n}\left[v_i\left(n_{Di} - 1\right)\right] \tag{2}$$

Eykman:

$$\frac{n_{Dm}^2 - 1}{n_{Dm}^2 + 0.4} = \sum_{i=1}^{n}\left[v_i\left(\frac{n_{D1}^2 - 1}{n_{Di}^2 + 0.4}\right)\right] \tag{3}$$

Newton:

$$n_{Dm}^2 - 1 = \sum_{i=1}^{n}\left[v_i\left(n_{Di}^2 - 1\right)\right] \tag{4}$$

Kay:

$$n_{Dm} = v_1 n_{D1} + v_2 n_{D2} \tag{5}$$

where n_{Dm} is the refractive index of the mixture and n_{Di} and v_i are the refractive index and the volumetric fraction of component i, respectively.

The accuracy of predictive models was estimated with the root mean square prediction difference (RMSPD).

$$RMSPD = 100\sqrt{\frac{1}{n}\sum_{i=1}^{n}\left[\frac{Y_{cal,i} - Y_{\exp,i}}{Y_{\exp,i}}\right]^2} \tag{6}$$

where Y_{cal}, Y_{exp} are the calculated and experimental values, respectively, and n is the number of experimental data.

The experimental values of the refractive index for the mixtures of biodiesel + diesel fuel and biodiesel + toluene at various temperatures and the result of predictive calculation are presented in Table 2 and 3.

From Table 2 it can be seen that the refractive index of the three types of biodiesel made from rapeseed oil differs from each other by ±0.0008. The refractive index of toluene is greater, and the refractive index of diesel fuel is lower than the refractive index of biodiesel. The three types of biodiesel mixtures present close values for refractive index, although the initial values of pure components differ.

Table 2

Refractive indices of binary mixtures at different temperatures

v_1	Bio_I+Diesel Fuel	Bio_I+Tol	Bio_{II}+Diesel fuel	Bio_{II}+Tol	Bio_{III}+Diesel fuel	Bio_{III}+Tol
			T=298.15K			
0.00	1.4650	1.4937	1.4650	1.4937	1.4650	1.4937
0.05	1.4645	1.4915	1.4645	1.4914	1.4644	1.4914
0.10	1.4640	1.4890	1.4640	1.4889	1.4639	1.4888
0.15	1.4635	1.4872	1.4634	1.4870	1.4633	1.4869
0.20	1.4629	1.4853	1.4628	1.4851	1.4626	1.4850
0.25	1.4624	1.4835	1.4623	1.4833	1.4621	1.4832
0.30	1.4619	1.4816	1.4618	1.4814	1.4616	1.4813
0.35	1.4613	1.4797	1.4612	1.4795	1.4610	1.4794
0.40	1.4608	1.4779	1.4606	1.4776	1.4604	1.4775
0.45	1.4603	1.4758	1.4601	1.4755	1.4598	1.4754
0.50	1.4597	1.4737	1.4595	1.4734	1.4592	1.4733
0.55	1.4592	1.4719	1.4590	1.4716	1.4587	1.4714
0.60	1.4587	1.4698	1.4585	1.4695	1.4582	1.4693
0.65	1.4582	1.4681	1.4580	1.4678	1.4576	1.4676
0.70	1.4577	1.4662	1.4574	1.4658	1.4570	1.4656
0.75	1.4572	1.4640	1.4569	1.4637	1.4565	1.4634
0.80	1.4567	1.4619	1.4564	1.4618	1.4560	1.4612
0.85	1.4562	1.4601	1.4559	1.4599	1.4554	1.4594
0.90	1.4556	1.4583	1.4554	1.4581	1.4548	1.4576
0.95	1.4552	1.4565	1.4549	1.4563	1.4544	1.4558
1.00	1.4548	1.4548	1.4545	1.4545	1.4540	1.4540
			T=303.15K			
0.00	1.4631	1.4905	1.4631	1.4905	1.4631	1.4905
0.05	1.4626	1.4886	1.4626	1.4885	1.4625	1.4885
0.10	1.4622	1.4866	1.4621	1.4865	1.4620	1.4864
0.15	1.4617	1.4847	1.4616	1.4845	1.4614	1.4844
0.20	1.4611	1.4827	1.4610	1.4825	1.4608	1.4823
0.25	1.4606	1.4810	1.4605	1.4808	1.4603	1.4806
0.30	1.4600	1.4792	1.4599	1.4790	1.4597	1.4788
0.35	1.4595	1.4774	1.4594	1.4772	1.4591	1.4770
0.40	1.4590	1.4756	1.4589	1.4755	1.4585	1.4752
0.45	1.4585	1.4737	1.4583	1.4735	1.4579	1.4732
0.50	1.4579	1.4718	1.4578	1.4715	1.4573	1.4712
0.55	1.4574	1.4699	1.4573	1.4696	1.4568	1.4692
0.60	1.4569	1.4679	1.4567	1.4675	1.4562	1.4671
0.65	1.4564	1.4661	1.4562	1.4657	1.4557	1.4653
0.70	1.4559	1.4642	1.4556	1.4639	1.4551	1.4634
0.75	1.4554	1.4622	1.4551	1.4619	1.4546	1.4613
0.80	1.4549	1.4601	1.4545	1.4600	1.4540	1.4591
0.85	1.4544	1.4585	1.4540	1.4582	1.4534	1.4575
0.90	1.4539	1.4569	1.4534	1.4566	1.4528	1.4559
0.95	1.4533	1.4549	1.4529	1.4545	1.4522	1.4538
1.00	1.4528	1.4528	1.4524	1.4524	1.4516	1.4516

			T=313.15K			
0.00	1.4591	1.4850	1.4591	1.4850	1.4591	1.4850
0.05	1.4587	1.4831	1.4587	1.4830	1.4586	1.4830
0.10	1.4582	1.4812	1.4582	1.4811	1.4581	1.4810
0.15	1.4578	1.4794	1.4577	1.4793	1.4576	1.4791
0.20	1.4573	1.4776	1.4572	1.4775	1.4570	1.4772
0.25	1.4568	1.4758	1.4566	1.4756	1.4564	1.4753
0.30	1.4563	1.4739	1.4561	1.4737	1.4558	1.4733
0.35	1.4558	1.4723	1.4556	1.4721	1.4553	1.4717
0.40	1.4553	1.4706	1.4551	1.4704	1.4547	1.4700
0.45	1.4548	1.4689	1.4546	1.4686	1.4542	1.4682
0.50	1.4543	1.4672	1.4541	1.4669	1.4536	1.4664
0.55	1.4539	1.4653	1.4536	1.4649	1.4531	1.4644
0.60	1.4534	1.4634	1.4531	1.4631	1.4525	1.4624
0.65	1.4529	1.4617	1.4525	1.4613	1.4519	1.4606
0.70	1.4524	1.4599	1.4520	1.4596	1.4514	1.4588
0.75	1.4519	1.4583	1.4515	1.4579	1.4508	1.4571
0.80	1.4514	1.4566	1.4510	1.4562	1.4503	1.4554
0.85	1.4509	1.4550	1.4505	1.4545	1.4497	1.4537
0.90	1.4505	1.4533	1.4500	1.4528	1.4492	1.4520
0.95	1.4500	1.4514	1.4495	1.4509	1.4486	1.4500
1.00	1.4495	1.4495	1.4490	1.4490	1.4480	1.4480
			T=323.15K			
0.00	1.4548	1.4790	1.4548	1.4790	1.4548	1.4790
0.05	1.4544	1.4772	1.4544	1.4772	1.4543	1.4771
0.10	1.4540	1.4754	1.4539	1.4753	1.4538	1.4752
0.15	1.4535	1.4737	1.4534	1.4736	1.4533	1.4734
0.20	1.4530	1.4720	1.4528	1.4720	1.4527	1.4716
0.25	1.4525	1.4703	1.4523	1.4702	1.4521	1.4698
0.30	1.4521	1.4686	1.4518	1.4685	1.4516	1.4680
0.35	1.4516	1.4671	1.4513	1.4669	1.4510	1.4665
0.40	1.4511	1.4656	1.4508	1.4653	1.4505	1.4650
0.45	1.4506	1.4638	1.4502	1.4635	1.4499	1.4631
0.50	1.4501	1.4620	1.4497	1.4616	1.4494	1.4612
0.55	1.4496	1.4604	1.4492	1.4600	1.4488	1.4596
0.60	1.4491	1.4588	1.4487	1.4584	1.4483	1.4580
0.65	1.4486	1.4570	1.4482	1.4566	1.4477	1.4561
0.70	1.4481	1.4552	1.4477	1.4547	1.4472	1.4542
0.75	1.4477	1.4534	1.4472	1.4529	1.4467	1.4523
0.80	1.4472	1.4515	1.4467	1.4512	1.4461	1.4503
0.85	1.4467	1.4500	1.4462	1.4495	1.4456	1.4488
0.90	1.4462	1.4484	1.4457	1.4479	1.4450	1.4472
0.95	1.4457	1.4468	1.4451	1.4462	1.4444	1.4455
1.00	1.4452	1.4452	1.4446	1.4446	1.4438	1.4438

Table 3

Root mean square prediction difference (RMSPD) as resulted for predictive models at different temperatures

System	T, K	Model				
		Lorentz-Lorenz	Dale-Gladstone	Eykman	Newton	Kay
		RMSPD %	RMSPD %	RMSPD %	RMSPD %	RMSPD %
Bio_I + Diesel fuel	*298.15*	0.0083	0.0087	0.0078	0.0090	0.0087
	303.15	0.0039	0.0038	0.0042	0.0037	0.0038
	313.15	0.0046	0.0043	0.0049	0.0040	0.0043
	323.15	0.0073	0.0069	0.0077	0.0066	0.0069
Bio_I + Tol	*298.15*	0.0257	0.0314	0.0198	0.0369	0.0314
	303.15	0.0132	0.0100	0.0191	0.0102	0.0100
	313.15	0.0117	0.0151	0.0105	0.0192	0.0151
	323.15	0.0109	0.0129	0.0114	0.0161	0.0129
Bio_{II} + Diesel fuel	*298.15*	0.0103	0.0107	0.0097	0.0111	0.0107
	303.15	0.0038	0.0035	0.0043	0.0022	0.0035
	313.15	0.0042	0.0040	0.0045	0.0038	0.0040
	323.15	0.0046	0.0043	0.0051	0.0041	0.0043
Bio_{II} + Tol	*298.15*	0.0305	0.0367	0.0236	0.0425	0.0367
	303.15	0.0131	0.0116	0.0181	0.0132	0.0116
	313.15	0.0127	0.0168	0.0102	0.0213	0.0168
	323.15	0.0089	0.0115	0.0094	0.0153	0.0115
Bio_{III} + Diesel fuel	*298.15*	0.0131	0.0136	0.0125	0.0141	0.0136
	303.15	0.0040	0.0036	0.0045	0.0034	0.0036
	313.15	0.0057	0.0052	0.0063	0.0048	0.0052
	323.15	0.0068	0.0063	0.0074	0.0059	0.0063
Bio_{III} + Tol	*298.15*	0.0292	0.0350	0.0231	0.0408	0.0350
	303.15	0.0157	0.0135	0.0211	0.0142	0.0135
	313.15	0.0155	0.0196	0.0128	0.0242	0.0196
	323.15	0.0140	0.0169	0.0130	0.0206	0.0169

The refractive index of mixtures with biodiesel, diesel fuel and toluen decreases with the increasing of biodiesel content, and with temperature increasing.

The RMSPD errors for the five predictive models: Lorentz-Lorentz, Dale-Gladstone, Eykman, Newton and Kay are presented in Table 3. As can be seen from Table 3 there are no significant differences regarding the accuracy of prediction for refractive indices of mixtures using Eq.1-5, the accuracy being very good. To note the accuracy of prediction for biodiesel blends with diesel fuel. The refractive index of mixtures can be predicted with a good accuracy using different mixing rules.

The correlation of refractive index with composition and temperature

From the experimental data the correlation equations of refractive index with composition and temperature were obtained. The equations can be used to estimate the refractive index of biodiesel mixtures of different degree of blending at any temperature in the studied domain.

The correlation equation was developed using the model proposed by Krisnangkura [21] for biodiesel properties:

$$\boldsymbol{ln\, n_{Dm} = a + b \cdot v_1 + \frac{c}{T} + \frac{d \cdot v_1}{T}} \quad (7)$$

where a, b, c, and d are correlation constants, T is absolute temperature and $\boldsymbol{v_1}$ is volume fraction of biodiesel.

The particular equations for mixtures of biodiesel obtained from rapeseed oil with diesel fuel and toluene (Tol) are presented in Table 4.

Table 4

Predictive equations for refractive index of binary biodiesel mixtures

System	Equation	T, K	RMSPD %
Bio_I + Diesel fuel	$\ln n_{Dm} = 0.2938 - 0.0008v_1 + \frac{26.303}{T} - \frac{1.8985v_1}{T}$	298.15	0.01
		303.15	0.01
		313.15	0.02
		323.15	0.02
Bio_I + Tol	$\ln n_{Dm} = 0.2783 + 0.0147v_1 + \frac{36.945}{T} - \frac{12.323v_1}{T}$	298.15	0.11
		303.15	0.08
		313.15	0.07
		323.15	0.12
Bio_{II} + Diesel fuel	$\ln n_{Dm} = 0.2934 - 0.0033v_1 + \frac{26.424}{T} - \frac{1.2791v_1}{T}$	298.15	0.01
		303.15	0.01
		313.15	0.03
		323.15	0.02
Bio_{II} + Tol	$ln\, n_{Dm} = 0.2795 + 0.0101v_1 + \frac{36.548}{T} - \frac{11.091v_1}{T}$	298.15	0.09
		303.15	0.07
		313.15	0.06
		323.15	0.10
Bio_{III} + Diesel fuel	$ln\, n_{Dm} = 0.2939 - 0.0063v_1 + \frac{26.260}{T} - \frac{0.5733v_1}{T}$	298.15	0.02
		303.15	0.02
		313.15	0.04
		323.15	0.02
Bio_{III} + Tol	$ln\, n_{Dm} = 0.2779 + 0.0088v_1 + \frac{36.956}{T} - \frac{10.742v_1}{T}$	298.15	0.07
		303.15	0.06
		313.15	0.05
		323.15	0.08

The accuracy of the prediction with the proposed model (Eq.7) is evaluated by the root mean square prediction difference (RMSPD) (Table 4). The measured values and predicted values are in good agreement and they are favorable for practical applications. The maximum error registered was 0.12%.

The correlations between refractive index and density or viscosity

Experimental data of refractive index can be used to calculate other properties of mixtures more difficult to measure in practice.

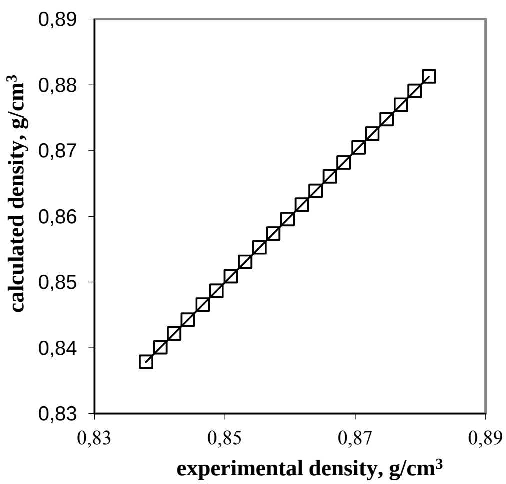
Correlative equations for density and viscosity calculation from the refractive index of biodiesel + diesel fuel mixtures were proposed in our previous paper [13]. The models were tested on the experimental data presented in Table 2. The particular equations obtained for the biodiesel + diesel fuel mixtures at 298.15 K are presented in Table 5. The equations represent well the relationship betwen density and viscosity, respectively, and the refractive index.

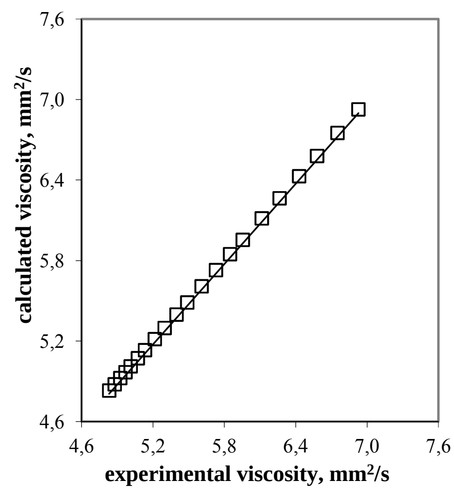
Table 5

Correlative equations between refractive index and density or viscosity for biodiesel + diesel fuel mixtures at 298.15 K

System	T, K	Equation
Bio_I + Diesel fuel	298.15 K	$\eta_m = 15886 \cdot n_{Dm}^2 - 46583 \cdot n_{Dm} + 34154$ (8) $d_m = -4.1924 \cdot n_{Dm} + 6.9796$ (9)
Bio_{II} + Diesel fuel	298.15 K	$\eta_m = 18608 \cdot n_{Dm}^2 - 54522 \cdot n_{Dm} + 39942$ (10) $d_m - 3.6416 \cdot n_{Dm} + 6.1725$ (11)
Bio_{III} + Diesel fuel	298.15 K	$\eta_m = 11429 \cdot n_{Dm}^2 - 33497 \cdot n_{Dm} + 24550$ (12) $d_m = -3.6503 \cdot n_{Dm} + 6.1854$ (13)

For example, Fig. 2 shows comparatively the experimental and calculated data of density and viscosity for $biodiesel_I$ + diesel fuel binary mixtures. The points signs represent the calculated density and viscosity values and the lines, the measured values, ones.





a) b)

Fig 2. Calculated density with Eq.8 *versus* experimental density (a) and calculated viscosity with Eq.9 *versus* experimental viscosity (b) for binary mixture bio_I + diesel fuel:
─ calculated values; □ experimental data.

The RMSPD values of density correlation are 0.0296 % in the case of bio_I + diesel fuel mixtures, 0.0445 % in the case of bio_{II} + diesel fuel mixtures and 0.0506 % in the case of bio_{III} + diesel fuel mixtures. The RMSPD values of viscosity correlation are 0.5262 % in the case of bio_I + diesel fuel mixtures, 11.93 % in the case of bio_{II} + diesel fuel mixtures and 22.97 % in the case of bio_{III} + diesel fuel mixtures. The accuracy of density prediction based on refractive index of binary mixtures biodiesel + diesel fuel is very good. For the viscosity, the results show that the equations can be used for bio_I + diesel fuel system and for other systems, can be only estimative.

4. Conclusions

Experimental refractive index were measured for biodiesel + diesel fuel and biodiesel + toluene mixtures using three types of biodiesel obtained by transesterification of rapeseed oils.

The experimental data were used to obtain predictive equations for biodiesel blends. Equations for refractive index versus composition and temperature for all mixtures have been derived. Also, the correlative equations for the viscosity or density of biodiesel + diesel fuel mixtures with refractive index are proposed.

The experimental data were used to test the prediction capability of Lorentz-Lorentz, Dale-Gladstone, Eykman, Newton and Kay models. The mixing rules can predict with a good accuracy the refractive indices of studied mixtures.

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