

MEASUREMENTS AND CORRELATIONS OF THE VISCOSITY OF ISOPROPANOL MIXTURES WITH DIESEL FUEL AND BIODIESEL

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ABSTRACT. Isopropanol could be an additive or substitute for diesel fuel due to some of its key properties. Viscosity influences the atomization and spray formation, affecting the combustion process and the emission of diesel engine. Experimental data of viscosity of diesel fuel + isopropanol and biodiesel + isopropanol mixtures over the entire composition range at five temperatures (288.15, 293.15, 303.15, 313.15 and 323.15 K) are presented. Based on experimental data, the ability of predictive and correlative models to estimate the viscosity of the studied systems was tested. The best accuracy in viscosity estimation at a desired temperature is given by a polynomial equation, followed by Grunberg-Nissan equation. The value of Grunberg-Nissan interaction parameter G_{12} increases with temperature increasing, following a trend described by a polynomial equation. Negative values for deviation in viscosity were obtained for diesel fuel + isopropanol mixtures and positive values for biodiesel + isopropanol mixtures. The best fit with viscosity experimental data for viscosity dependence on temperature was provided by Vogel model.

Keywords: *viscosity, biodiesel, isopropanol, diesel, viscosity models*

INTRODUCTION

The search for sustainable and alternative fuels is a major topic of today research, taking into account that the use of fossil fuels is an important source for environmental pollution. It was proved that oxygenated fuels improve the combustion process, reducing harmful emissions like greenhouse gas and particulate matter [1]. The effect of biodiesel and alcohols such as

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methanol, ethanol and butanol addition to diesel fuel, on blends properties and diesel engine behaviour, respectively, have been widely investigated [2-5] due to their renewable nature and improved combustion. The need to diversify the biofuels used as additives or substitutes for fossil fuels is a stringent necessity taking into account the continuously increasing demand for energy in transport sector. Isopropanol could be a promising fuel alternative for emission control of combustion of fuel blends in compression engines. It was proved that isopropanol together with ethanol and n-butanol can be produced by fermentation of sugars from lignocellulosic biomass in the presence of appropriate bacteria [6].

The interest for isopropanol as additive or substitute for diesel fuel is due to some of its key properties that overcome methanol and ethanol like higher cetane number and energy content, reduced corrosivity due to its less water-absorbing nature, greater miscibility with diesel fuel, more like diesel fuel viscosity and density [7]. The addition of isopropanol to diesel fuel was proved to reduce the harmfully emissions of diesel engine [8]. Some properties of diesel fuel and biodiesel, as density and viscosity, important for diesel engine operation, can be improved by isopropanol addition. Experimental and correlated data of these properties for diesel + isopropanol and biodiesel + isopropanol mixtures are rarely reported in the literature. Density experimental data together with calculated excess molar volume values of pseudo-binary diesel fuel and biodiesel with isopropanol mixtures and corresponding ternary mixture, have been reported [9, 10]. As far as we know, in the literature have been presented data on viscosity only for the ternary diesel + biodiesel + isopropanol mixture [5].

Viscosity which expresses the internal resistance or friction to flow of a fluid, influences the atomization process and spray formation in accordance with the characteristics of the injection system of diesel engine [11], affecting the combustion process, and the exhaust emission composition, respectively. As viscosity is an important parameter to characterize fuels used for diesel engine, the main aim of the current study which is a continuation of our experimental work on properties of blends of fossil fuels with biofuels [10, 12], is to report experimental viscosity data of pseudo-binary mixtures of diesel fuel and biodiesel with isopropanol over the entire composition range at 288,15 K to 323,15 K. The effects of alcohol fraction and temperature on the viscosity of diesel fuel + isopropanol and biodiesel + isopropanol systems have been investigated. The accuracy of different equations to estimate the viscosity of these systems with isopropanol have been assessed. An equation for the dependency on temperature of G_{12} interaction parameter from Grunberg – Nissan model is proposed. Deviation in viscosity ($\Delta\eta$) was obtained based on viscosity data. The results are useful for modelling diesel

engine behaviour fuelled by such blends in order to identify the best blend composition and to optimise engine design, resulting in reduced environment pollution.

MODELS FOR VISCOSITY ESTIMATION

Viscosity of a liquid mixture at atmospheric pressure depends on mixture composition and temperature. Various models proposed in the literature to correlate the experimental viscosity of liquid mixtures were tested for diesel fuel with biofuel blends. Mixing rules like simplified Grunberg-Nissan [13], Kendall-Monroe [14] and Frenkel [15] predict the viscosity of liquid mixtures from the viscosity of mixture components at a desired temperature, without the need of adjustable parameters (Table 1). Other correlative-predictive or correlative models with parameter were used to calculate the viscosity of diesel fuel blends with biofuels: Grunberg-Nissan [13] and polynomial type [16], respectively (Table 1). Exponential type temperature dependent equations: Andrade [17], Vogel [18] and Tat and Van Gerpen [19] were used to estimate the viscosity of different diesel fuel with biofuel blends (Table 1). The significance of symbols used in Eqs. (1-8) is as follows: η represents the dynamic viscosity of the mixture, η_1 , η_2 and x_1 , x_2 represent the dynamic viscosity and mole fraction, respectively for the components of the mixture, T represents the absolute temperature, G_{12} , a , b , c , A , B , C are adjustable parameters.

Table 1. Models for viscosity calculation

Model	Ref.	Equation
Viscosity dependence on composition		
Grunberg-Nissan simplified	[12]	$\eta = \exp(x_1 \ln \eta_1 + x_2 \ln \eta_2)$ (1)
Kendall-Monroe	[13]	$\eta = (x_1 \eta_1^{1/3} + x_2 \eta_2^{1/3})^3$ (2)
Frenkel	[14]	$\eta = \exp(x_1^2 \ln \eta_1 + x_2^2 \ln \eta_2 + 2x_1 x_2 \ln \eta_{12})$ (3) where $\eta_{12} = 0.5\eta_1 + 0.5\eta_2$
Grunberg-Nissan	[12]	$\eta = \exp(x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_1 x_2 G_{12})$ (4)
Polynomial	[15]	$\eta = ax^2 + bx + c$ (5)
Viscosity dependence on temperature		
Andrade	[16]	$\eta = A \exp(B/T)$ (6)
Vogel	[17]	$\eta = \exp(A + B/(C + T))$ (7)
Tat and Van Gerpen	[18]	$\eta = \exp(A + B/T + C/T^2)$ (8)

The ability of different models to accurately predict the viscosity of fuels mixtures can be assessed by the means of statistical indicators like average relative deviation (ARD %) and standard deviation (SD). The following equations were used:

$$ARD(\%) = \frac{100}{N} \sum_{i=1}^N \left(\frac{|\eta_{xp,i} - \eta_{cal,i}|}{\eta_{exp,i}} \right) \quad (9)$$

$$SD = \left(\frac{\sum_{i=1}^N (\eta_{exp,i} - \eta_{cal,i})^2}{N-m} \right)^{1/2} \quad (10)$$

where η_{exp} and η_{cal} represent the experimental and calculated viscosity, respectively, N represents the number of experimental points, and m is the number of model parameters.

RESULTS AND DISCUSSION

Experimental data of dynamic viscosity of diesel fuel + isopropanol and biodiesel + isopropanol mixtures in the temperature range of 288.15 K to 323.15 K are presented in Table 2.

Table 2. Experimental data of viscosity of diesel fuel + isopropanol and biodiesel + isopropanol mixtures

Mole fraction (x_2)	Dynamic viscosity (mPa·s)				
	288.15 K	293.15 K	303.15 K	313.15 K	323.15 K
Diesel fuel (1) + isopropanol (2)					
0.0000	4.0629	3.5437	2.7978	2.2529	1.8505
0.1002	3.8959	3.4019	2.6495	2.1461	1.7952
0.1997	3.6824	3.2163	2.5431	2.0526	1.7010
0.3972	3.3610	2.8982	2.3201	1.8659	1.5322
0.5982	3.1501	2.7482	2.1358	1.6932	1.3865
0.7970	2.9045	2.5375	1.9451	1.5184	1.2218
0.9086	2.8412	2.4288	1.8470	1.4216	1.1254
1.0000	2.8999	2.4633	1.8358	1.3825	1.0514
Biodiesel (1) + isopropanol (2)					
0.0000	7.5205	6.4987	5.0198	4.0301	3.2988
0.1021	7.1280	6.1997	4.8583	3.8611	3.1252
0.2019	6.4882	5.6214	4.4765	3.4920	2.8690
0.3984	5.7949	5.0505	3.9295	3.1726	2.5511
0.6027	4.9749	4.3562	3.3849	2.6926	2.1919
0.7987	3.9183	3.3453	2.5928	2.0017	1.6374
0.9301	3.1481	2.7238	2.0874	1.6164	1.2820
1.0000	2.8999	2.4633	1.8358	1.3825	1.0514

The viscosity of isopropanol is lower than that of diesel fuel and biodiesel, respectively. The decrease of dynamic viscosity of diesel fuel + isopropanol and biodiesel + isopropanol pseudo-binary systems with alcohol concentration increasing and temperature increasing can be observed as expected. The decreasing trend of viscosity with alcohol content increasing is higher for biodiesel + isopropanol system. The addition of isopropanol to biodiesel brings the viscosity of the resulting mixture closer to that of diesel fuel. It can be observed that the decrease of the viscosity of diesel fuel + isopropanol mixtures with the increasing alcohol content shows a minimum point in the concentration zone of $x = 0.9$ for temperature ranging from 288.15 K to 303.15 K. The minimum reduces with temperature increasing, to disappear for temperatures greater than 303.15 K.

Viscosity – composition correlation

Equations (1-5) (Table 1) were used to calculate the dynamic viscosity of diesel fuel + isopropanol and biodiesel + isopropanol mixtures for temperature ranging from 288.15 K to 323.15 K. Based on experimental data of dynamic viscosity, the fitting parameters of Eq. (4) and Eq. (5) were calculated (Table 3). The obtained values for interaction parameter G_{12} are greater for biodiesel + isopropanol mixture, compared to diesel fuel + isopropanol mixture. The same observation was reported in the literature on biodiesel and diesel fuel blends with ethanol and n-butanol [4].

Table 3. Fitting parameters of Grunberg-Nissan (Eq.4) and polynomial type (Eq.5) models and corresponding accuracies (SD)

Parameter	Temperature (K)				
	288.15	293.15	303.15	313.15	323.15
Diesel fuel + isopropanol					
Eq. (4)					
G_{12}	-0.2593	-0.2090	-0.1061	0.0071	0.1895
SD (mPa·s)	0.0521	0.0399	0.0209	0.0112	0.0090
Eq. (5)					
a	0.9675	0.7460	0.3604	0.1579	-0.0225
b	-2.1997	-1.8706	-1.3390	-1.0409	-0.7849
c	4.0839	3.5580	2.7926	2.2525	1.8591
SD (mPa·s)	0.0402	0.0367	0.0184	0.0100	0.0097
Biodiesel + isopropanol					
Eq. (4)					
G_{12}	0.5352	0.6033	0.7512	0.8303	0.9642
SD (mPa·s)	0.1295	0.1232	0.0823	0.0983	0.1018
Eq. (5)					
a	-0.6842	-0.8085	-0.8950	-0.8168	-0.7515
b	-3.9247	-3.2208	-2.3170	-1.8105	-1.4421
c	7.4763	6.4673	5.0356	4.0081	3.2658
SD (mPa·s)	0.1052	0.1017	0.0559	0.0684	0.0501

It can be observed (Table 3) that the values of G_{12} parameters increase with temperature increasing, following a trend described by polynomial equations:

$$G_{12,d} = 0.0002T^2 - 0.1086T + 14.5130 \quad (R^2 = 0.9908) \quad (11)$$

$$G_{12,bd} = 0.00002T^2 + 0.0033T - 1.6791 \quad (R^2 = 0.9680) \quad (12)$$

where $G_{12,d}$ refers to diesel fuel + isopropanol system and $G_{12,bd}$ refers to biodiesel + isopropanol system.

The value of parameter G_{12} was also determined for the temperature of 313.15 K (the reference temperature in accordance with fuels quality standards) following the equations proposed by Lapuerta et al. [4]:

$$G_{12,d} = 0.11n^2 - 1.242n + 2.897 \quad (13)$$

$$G_{12,bd} = 0.141n^2 - 1.486n + 3.851 \quad (14)$$

where n represents the number of carbon atoms from the alcohol molecule. The resulting values were $G_{12,d} = 0.161$ and $G_{12,b} = 0.662$, respectively. It can be observed that these values are not very close to the values obtained based on experimental data but, it should be noticed that Eqs. (13) and (14) have been proposed for linear chain alcohols.

The accuracy of Eqs. (1-5) to estimate the viscosity of diesel fuel + isopropanol and biodiesel + isopropanol mixtures over the temperature range of 288.15 K to 323.15 K, expressed by ARD (%) is presented in Figure 1. The best results (smaller ARD values) have been obtained for diesel fuel + isopropanol system.

Between the mixing rules, Eq. (1) shows the best results in predicting the viscosity of diesel fuel + isopropanol mixtures (ARD < 3.4%), and Eq. (3) for biodiesel + isopropanol mixtures (ARD < 7.69 %). Mixing rules (Eqs. 1 - 3) have lower accuracy compared to equations with parameters (Eqs. 4 and 5). As presented in Fig.1, the best accuracy in viscosity estimation at a desired temperature is given by Eq. (5) (ARD < 0.89 % for diesel fuel + isopropanol system and ARD < 1.41 % for biodiesel + isopropanol system), followed by Eq. (4) (ARD < 1.21 % for isopropanol mixtures with diesel fuel and ARD < 3.48 % for isopropanol mixtures with biodiesel). Comparing the results obtained with Grunberg-Nissan simplified model (Eq. 1) with Grunberg-Nissan model (Eq. 4), it can be observed that the application of interaction parameter G_{12} improves the result for both studied systems with isopropanol. Similar results were reported in the literature for blends of diesel fuel with ethanol and n-butanol, and biodiesel with ethanol, n-propanol, and n-butanol [4]. This result is expected for complex systems like diesel fuel and biodiesel mixtures with

isopropanol, taking into account the structural differences between the alcohol molecule (highly polar) and the hydrocarbons molecules from diesel fuel (non-polar n-alkanes and reduced polarity aromatics) and fatty acid esters molecules from biodiesel (low polarity).

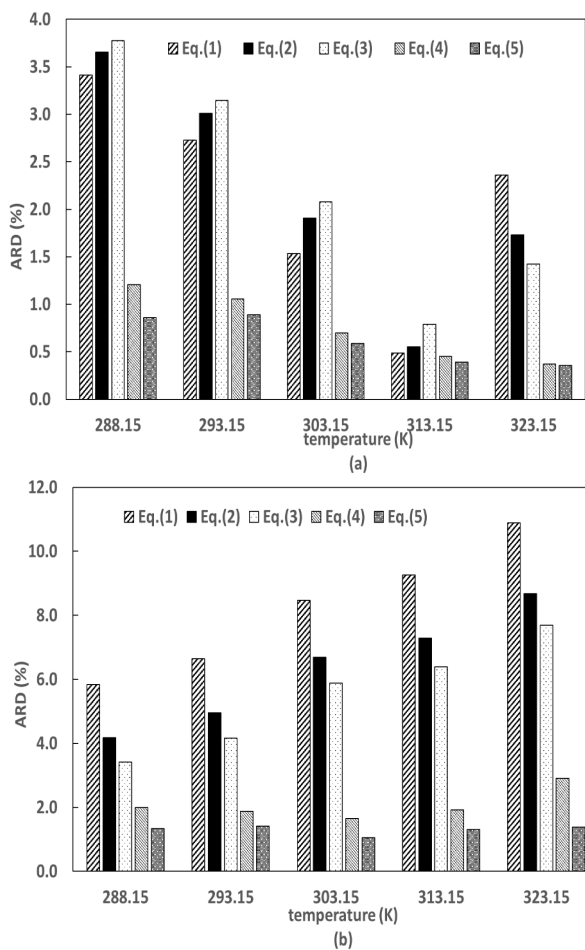


Figure 1. Accuracies (ARD %) of viscosity calculated with models described by Eqs.(1 - 5) for (a) diesel fuel + isopropanol and (b) biodiesel + isopropanol systems

Figure 2 presents dynamic viscosity calculated with Eqs. (1- 5), versus experimental viscosity for the reference temperature of 313.15 K. Eq. (4) was applied using G_{12} parameter value determined based on experimental data, and in accordance with Eqs. (13) and (14), respectively.

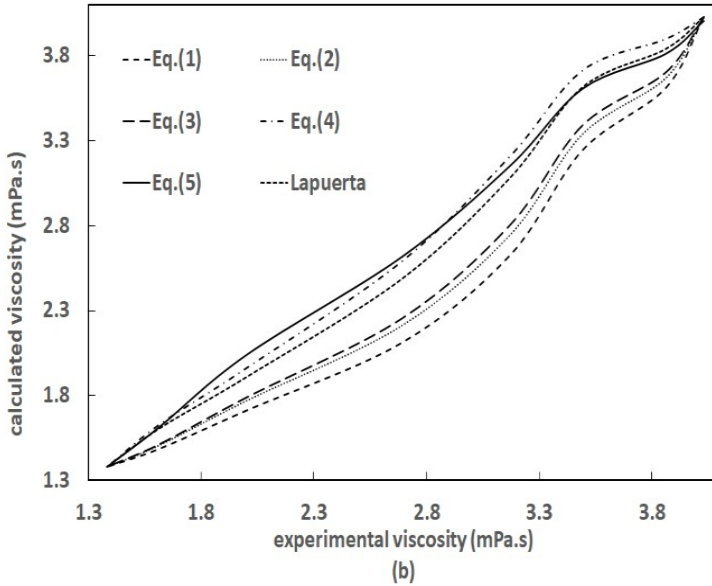
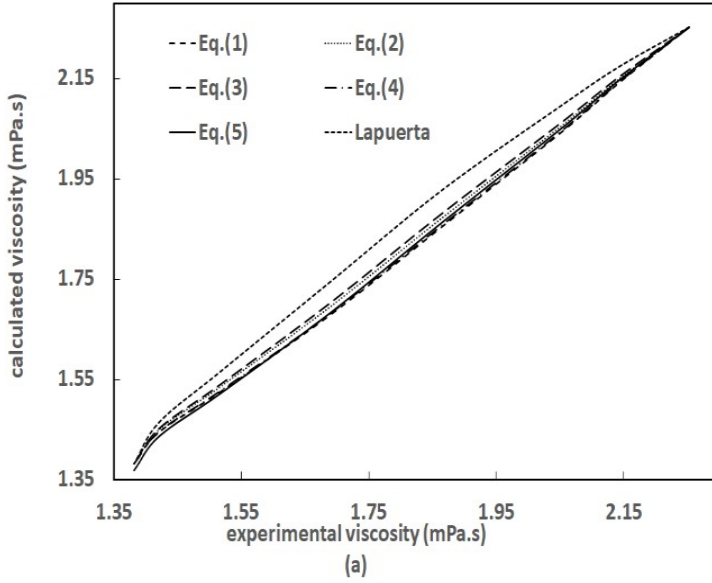


Figure 2. Calculated viscosity versus experimental viscosity for (a) diesel fuel + isopropanol and (b) biodiesel + isopropanol system at 313.15 K (G_{12} parameter value is calculated based on experimental data (Eq.4) and applying equations proposed by Lapuerta et al. [4])

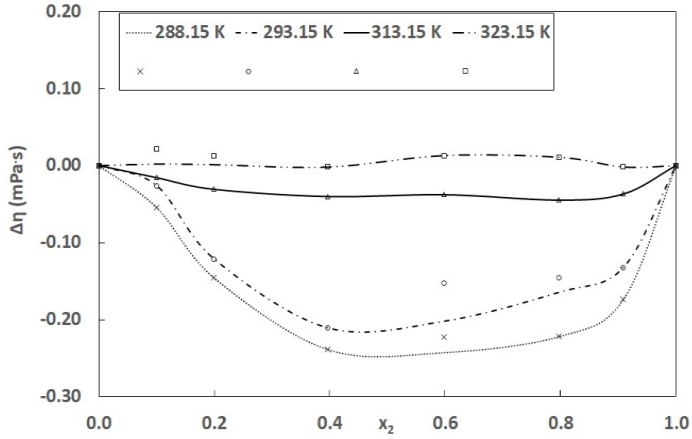
Viscosity estimation with Grunberg-Nissan model with the value of G_{12} parameter determined based on experimental data is more accurate than using the value determined based on Lapuerta equation [4] but, it must be noted that the accuracy of Grunberg-Nissan model using Lapuerta value for G_{12} parameter is greater than that corresponding to mixing rules (Eqs.1-3).

In order to evaluate the viscosity deviation at mixing ($\Delta\eta$) which is important in practice (production and equipment design), the following relation was used:

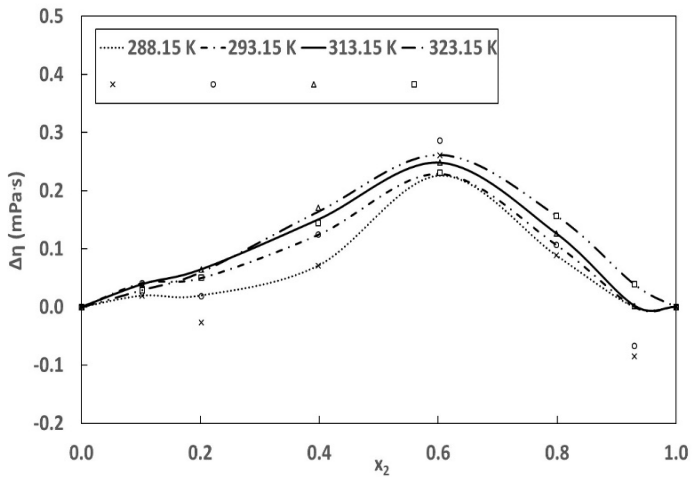
$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \quad (15)$$

Figure 3 presents deviation in viscosity variation with alcohol content of diesel fuel + isopropanol and biodiesel + isopropanol mixtures. The difference in the variation of $\Delta\eta$ with composition for diesel fuel + isopropanol and biodiesel + isopropanol mixtures can be influenced by molecular interactions, but also by the size and shape of mixtures components. Diesel components include linear and cyclic n-alkanes, olefins and aromatics, but non-polar n-alkane are predominant. Biodiesel is a mixture of monoalkyl esters of fatty acids with reduced polarity.

The $\Delta\eta$ as well as interaction parameter G_{12} are majority negative for diesel fuel + isopropanol mixtures indicating that specific interaction between unlike molecules is absent. Similar behaviour with diesel fuel + isopropanol system was reported in the literature for binary mixture of cyclopentane with isopropanol [19]. The $\Delta\eta$ as well as interaction parameter G_{12} are positive for biodiesel + isopropanol mixtures. Moderately positive values should indicate that specific interaction would be present between the functional groups of the components.



(a)



(b)

Figure 3. Deviation in viscosity ($\Delta\eta$) versus alcohol mole fraction for (a) diesel fuel + isopropanol and (b) biodiesel + isopropanol systems at 313.15 K (lines - smoothed data; points- calculated data)

Viscosity – temperature correlation

Equations (6-8) were applied to calculate the dynamic viscosity of diesel fuel + isopropanol and biodiesel + isopropanol mixtures for temperature range of 288.15 K to 323.15 K.

Table 4. Fitting parameters of Eqs. (6), (7) and (8) and standard deviations (SD) for diesel fuel + isopropanol mixtures

Parameter	x_2							
	0.0000	0.1002	0.1997	0.3972	0.5982	0.7970	0.9086	1.0000
Eq. (6)								
A	0.0029	0.0029	0.0029	0.0026	0.0016	0.0009	0.0006	0.0003
B	2086.86	2073.58	2060.29	2061.29	2191.12	2320.42	2453.40	2659.42
SD	0.0255	0.1879	0.2593	0.3655	0.4469	0.5155	0.5505	0.5775
Eq. (7)								
A	-3.1768	-5.7556	-5.7955	-5.9645	-6.4316	-6.9646	-7.4941	-8.3081
B	775.24	2019.16	2030.69	2058.80	2171.65	2310.79	2456.12	2715.16
C	-118.79	-3.8879	-1.7195	-0.7634	-1.3932	-0.3894	-0.2797	1.5381
SD	0.0109	0.0532	0.0330	0.0409	0.0206	0.0101	0.0164	0.0103
Eq. (8)								
A	-5.8459	-5.8361	-5.8304	-5.9785	-6.4611	-6.9723	-7.4997	-8.2633
B	2085.95	2069.82	2052.84	2068.20	2190.57	2316.05	2460.05	2687.79
C	13.6359	13.5377	13.4225	13.5176	14.3164	15.1317	16.0650	17.5347
SD	0.0945	0.0361	0.0603	0.0402	0.0236	0.0430	0.0062	0.0103

Table 5. Fitting parameters of Eqs. (6), (7) and (8) and standard deviations (SD) for biodiesel + isopropanol mixtures

Parameter	x_2							
	0.0000	0.1021	0.2019	0.3984	0.6027	0.7987	0.9301	1.0000
Eq. (6)								
A	0.0037	0.0036	0.0034	0.0030	0.0025	0.0011	0.0008	0.0003
B	2188.08	2188.48	2176.17	2176.76	2190.30	2346.59	2389.98	2659.41
SD	0.0772	0.0290	0.0488	0.0317	0.0183	0.0340	0.0051	0.0116
Eq. (7)								
A	-1.8004	-5.6588	-5.6805	-5.8003	-5.9924	-6.7652	-7.1499	-8.3081
B	486.81	2203.62	2175.79	2182.69	2189.00	2340.26	2389.72	2715.16
C	-160.61	1.1434	0.2523	0.9095	0.1349	0.0601	-0.0659	1.5381
SD	0.0122	0.0362	0.0603	0.0402	0.0233	0.0428	0.0061	0.0103
Eq. (8)								
A	-5.5886	-5.6304	-5.6729	-5.7773	-5.9865	-6.7597	-7.1511	-8.2633
B	2187.93	2186.70	2171.64	2169.13	2186.16	2336.06	2390.55	2687.79
C	14.3033	14.2867	14.1915	14.1726	14.2860	15.2748	15.6113	17.5347
SD	0.0945	0.0361	0.0603	0.0402	0.0236	0.0430	0.0062	0.0103

The values of the fitting parameters determined based on experimental viscosity data, and the corresponding standard deviations are listed in Table 4 and 5.

Comparison of the performance of Eqs. (6-8) used to calculate the dynamic viscosity of mixtures of diesel fuel and biodiesel with isopropanol at different temperatures can be made based on accuracy data (ARD %) presented in Figure 4. Similar good accuracies were obtained with Eqs. (6), (7), and (8), ARD (%) was no greater than 1.13 % for mixtures of isopropanol with diesel fuel, and no greater than 0.96 % for mixtures with biodiesel, but it

should be noted that Vogel model provides the best fit (ARD < 1.12 % for isopropanol mixtures with diesel fuel and ARD < 0.88 % for isopropanol mixtures with biodiesel). Vogel model can be recommended for dynamic viscosity calculation for diesel fuel + isopropanol and biodiesel + isopropanol mixtures.

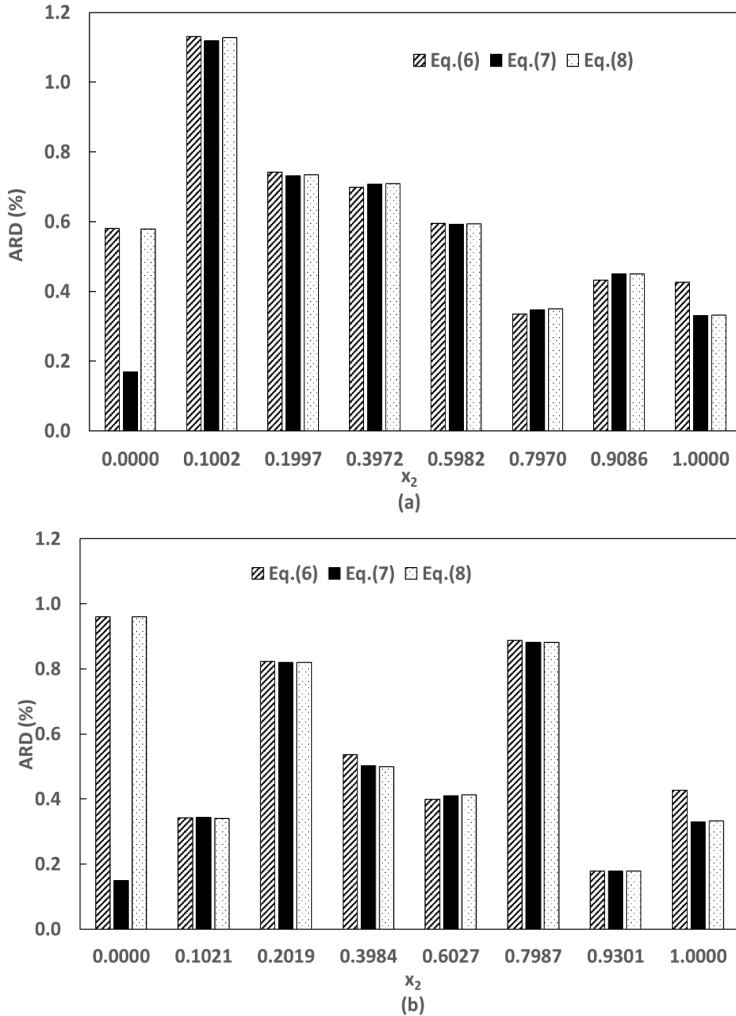


Figure 4. Deviation between calculated (Eqs. 6 - 8) and experimental dynamic viscosity for (a) diesel fuel + isopropanol and (b) biodiesel + isopropanol mixtures

CONCLUSIONS

Experimental measurements of dynamic viscosity of diesel fuel+ isopropanol and biodiesel + isopropanol systems over the entire composition range at 288,15 K, 293.15 K, 303.15 K, 313.15 K, and 323.15 K under atmospheric pressure have been reported. The addition of isopropanol to biodiesel helps to reduce the viscosity of biodiesel and to bring it closer to that of diesel fuel.

The ability of different models to estimate the viscosity of diesel fuel and biodiesel mixtures with isopropanol as a function of composition or temperature was assessed. A three parameters polynomial equation has the best accuracy of viscosity estimation at a given temperature for diesel fuel or biodiesel mixtures with isopropanol.

An equation is proposed to describe the dependency on temperature for the interaction parameter from Grunberg-Nissan model.

Positive values of $\Delta\eta$ have been obtained for diesel fuel + isopropanol mixtures and negative values for biodiesel + isopropanol mixtures. Among exponential equations used to estimate the viscosity of diesel fuel and biodiesel mixtures with isopropanol at different temperatures, Vogel model can be recommended.

EXPERIMENTAL SECTION

Diesel fuel used in this study was supplied by a local company and fulfils the requirements of the European Standard EN 590. Biodiesel obtained from a biodiesel producer follows the European Standard EN 14214 requirements. Table 6 shows properties of these two fuels. Isopropanol of at least 99.0% purity, contained 0.1 ppm of water and has an acidity value of 0.001 %. Pseudo-binary mixtures of diesel fuel with isopropanol, and biodiesel with isopropanol were prepared by mass using an analytical balance with a precision of $1 \cdot 10^{-4}$ g. In order to prevent evaporation, samples were kept in glass bottles tightly closed and filled at least of 90 % of the volume.

Viscosity of the investigated pseudo-binary systems was measured according to ASTM D 7042 method on an Anton Paar SVM 3000 viscometer at atmospheric pressure and at temperatures ranging from 288.15 K, to 323.15 K. The temperature of the samples was maintained by the integrated thermostated bath with an accuracy of ± 0.02 K. The viscometer was calibrated using ambient air and double distilled water before each series of measurements. The uncertainty in viscosity data was estimated to be ± 0.35 %.

Table 6. Properties of diesel fuel and biodiesel

Property	Method	Diesel fuel		Biodiesel	
		EN 590	exp*	EN 14214	exp*
Kinematic viscosity at 40°C (mm ² /s)	EN 3104	2.0 – 4.5	2.7393	3.50 – 5.00	4.6699
Density at 15°C (kg/m ³)	EN 12185	820 - 845	840.1	860 - 900	881.2
Water content (mg/kg)	EN 12937	max 200	97	max 500	259
Sulphur content (mg/kg)	ASTM D5453	max 10	8.2	max 10	5.0
Methylic esters of fatty acid (% wt)	EN 14103		0	min 96.5	98.2
Monglycerides (% wt)	EN 14105			max 0.80	
Diglycerides (% wt)	EN 14105			max 0.20	
Triglycerides (% wt)	EN 14105			max 0.20	
Mean molar mass (g/mol)			211		296
Cetane number	EN 4264	min 49	50	>51	52.8
Free glycerol (% wt)	EN 14105			max 0.02	0.015
Total glycerol (% wt)	EN 14105			max 0.25	0.23

*experimentally determined

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