

# Viscosity of Ternary Blends with Biodiesel, Diesel Fuel and Isopropanol at Different Temperatures

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*The aim of this study is to report experimental viscosity data of pseudo-ternary biodiesel + diesel fuel + isopropanol system covering the whole composition range and for temperature ranging from 293.15 K to 323.15 K. The accuracy of some equations to estimate the viscosity of this ternary system was evaluated.*

*Keywords: biodiesel, diesel fuel, isopropanol, viscosity, ternary mixture*

The environment pollution for which the transport sector is particularly responsible is one of the main concerns of our days. Biofuels like biodiesel and bioethanol are commonly used as replacement of fossil fuels, diesel fuel and gasoline respectively, in order to reduce the environment pollution. The need to diversify the biofuels used as additives or replacement for fossil fuels is a result of the continuous increasing fuels demand for internal combustion engines. Bioalcohols like isopropanol and other alcohols ( $C > 3$ ) could be an interesting option as blending components of diesel fuel and biodiesel, taking into account their advantages over methanol and ethanol like higher energy content, higher cetane index, higher miscibility with diesel fuel and reduced corrosivity [1]. Lower aliphatic alcohols (methanol and ethanol) studied as alternative fuels have solubility problems with diesel fuel [2]. Higher alcohols have come into attention due to the discovery of new convenient methods to obtain them from biomass with improved yield [1].

The lower viscosity of isopropanol obtained by bioresource technology compared to biodiesel recommends this bioalcohol to be blended with biodiesel in order to obtain more like diesel fuel properties allowing more biofuels quantities to be added to diesel fuel without the need of engine modification.

Physicochemical properties, including transport properties of pseudo-binary diesel fuel+biodiesel blends are widely investigated in literature [3-6]. Only a few studies are addressed to pseudo-ternary blends with diesel fuel and most of these investigated engine performance and exhaust emissions. Very few studies refer to properties such as viscosity and density of ternary fuel systems [7-8], the behavior of the engine and the impact on the environment being the most investigated aspects.

The addition of n-pentanol or n-hexanol to diesel+biodiesel blends results in the drastically reduction of the flash point and a decrease in the pour point and cloud point of the diesel+biodiesel+alcohol ternary blend was observed [9]. Regarding the emissions of a ternary blend containing 85 % v biodiesel, 5 % v diesel and 10 % v n-pentanol, a diminution in carbon monoxide was measured compared to pure diesel or biodiesel fuels [9]. The reduction amount of carbon monoxide and unburned hydrocarbons was registered too for a diesel engine fuelled with a ternary n-butanol+diesel+vegetable oil blend containing 60 % v alcohol [10]. It was shown that the

increasing alcohol content of a ternary diesel+palm oil+n-butanol blend results in reduced harmful emissions like nitrogen oxides, carbon monoxide, carbon dioxide and smoke [11]. An important decrease of nitrogen oxides, carbon monoxide and unburned hydrocarbons was registered for a ternary blend containing 65.5 % v diesel, 23.1 % v n-butanol and 11.4 % v cotton oil [12].

Viscosity is one of the important properties of fuels for diesel engine, affecting the pumping, spraying characteristics and combustion process. Continuing our studies on mixtures of fossil fuels with biofuels [13-16], the present paper reports viscosity data of pseudo-ternary biodiesel+diesel fuel+isopropanol system over the whole composition range and for temperatures between 293.15 K and 323.15 K. The ternary system is practically a pseudo-ternary mixture because both diesel fuel and biodiesel contain a mixture of the same class of chemical components, hydrocarbons in the case of diesel fuel and monoalkylesters of fatty acids for biodiesel. These fuels were characterised by physicochemical properties and average molar mass. The accuracy of some predictive and correlative equations to calculate the viscosity of this ternary system was evaluated. To the best of our knowledge, there are no properties data for this system in literature.

## Experimental part

Ternary biodiesel+diesel fuel+isopropanol mixtures were prepared at room temperature by weighing using an analytical balance with a precision of  $\pm 0.0001$  g. In order to cover the entire composition range, 19 mixtures were used for viscosity measurement. The samples were prepared into stoppered bottles filled at least of 90 % of volume in order to prevent evaporation.

Both diesel fuel without additives and the biodiesel used in this study fulfil the standard quality requirements, EN 590 for diesel fuel and EN 14214 for biodiesel, respectively. Physicochemical properties of these fuels are presented in table 1. Isopropanol was purchased by Merck and has a purity of 99.7% v/v. The molar mass of diesel fuel and biodiesel was determined using the cryoscopic method and also based on the chemical composition chromatographically determined for biodiesel (Clarus 500 GC chromatograph equipped with a FID detector and capillary column SGE BPX70 of high polarity; hydrogen flow rate of 20 mL/min). The resulted mean molar masses were

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**Table 1**  
PROPERTIES OF DIESEL FUEL AND BIODIESEL

Property	Method	Diesel fuel		Biodiesel	
		EN 590	experimental	EN 14214	experimental
Kinematic viscosity at 40 °C (mm <sup>2</sup> /s)	EN ISO 3104	2.0 – 4.5	2.6791	3.50 – 5.00	4.4189
Density at 15 °C (kg/m <sup>3</sup> )	EN ISO 12185	820 – 860	840.5	860 - 900	883.8
Water content (mg/kg)	EN ISO 12937	max 200	97	max 500	259
Sulfur content (mg/kg)	ASTM D5453	max 10	8.2	max 10	5
Cetane number	EN ISO 4264	min 49	50	>51	51
Flash point (°C)	EN ISO 2719	>55	64	>101	175
Methylic esters of fatty acids (% w/w)	EN 14103	5.0	0	min 96.5	98.9
Free glycerol (% w/w)	EN 14105			max 0.02	0.005
Total glycerol (% w/w)	EN 14105			max 0.25	0.155

**Table 2**  
FATTY ACID METHYL ESTER PROFILE OF BIODIESEL

fatty acid methyl ester <sup>+</sup>	C16:0	C18:0	C18:1	C18:2	C19:2	C20:0	C20:1	C20:2
% w/w	3.47	1.52	39.22	39.52	15.09	0.38	0.75	0.25

\*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.

of 210.01 g/mol for diesel fuel and of 295.18 g/mol for biodiesel. Table 2 presents the fatty acid methyl ester profile of biodiesel.

The experimental measurements of viscosity were carried out according to ASTM D 7042 method using the Anton Paar type SVM 3000 viscometer at atmospheric pressure and for temperatures ranging from 293.15 K, to 323.15 K at 5 K intervals. This equipment contains a cell for dynamic viscosity measurement, and a cell for density measurement. The kinematic viscosity value calculated by dividing the dynamic viscosity by the density is displayed at each temperature. The viscometer is equipped with a built-in constant temperature bath, the temperature stability being  $\pm 0.02$  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  $\pm 0.35$  %. All measurements were repeated three times, and the results were averaged.

### Equations

Mixing rules that predict and correlate the viscosity of fossil fuels with biofuels blends with composition or

$$\ln \eta = w_1 \ln \eta_1 + w_2 \ln \eta_2 + w_3 \ln \eta_3 \quad (1a)$$

$$\ln \eta = w_1 \ln \eta_1 + w_2 \ln \eta_2 + w_3 \ln \eta_3 + w_1 w_2 G_{12} + w_1 w_3 G_{13} + w_2 w_3 G_{23} \quad (1b)$$

$$\ln \eta = w_1 \ln \eta_1 + w_2 \ln \eta_2 + w_3 \ln \eta_3 + w_1 w_2 G_{12} + w_1 w_3 G_{13} + w_2 w_3 G_{23} + w_1 w_2 w_3 G_{123} \quad (1c)$$

b) Mc Allister equation [18] based on the theory of activated complex applied to the viscous flow phenomenon:

$$\begin{aligned} \ln \eta = & x_1^3 \ln(\eta_1 M_1) + x_2^3 \ln(\eta_2 M_2) + x_3^3 \ln(\eta_3 M_3) - \ln M_{avg} + 3x_1^2 x_2 \ln M_{12} + 3x_1^2 x_3 \ln M_{13} + \\ & 3x_2^2 x_3 \ln M_{23} + 3x_2 x_3^2 \ln M_{32} + 3x_1 x_3^2 \ln M_{31} + 3x_1^2 x_2 \ln M_{21} + 6x_1 x_2 x_3 \ln M_{123} + 3x_1^2 x_2 \ln \eta_{12} + \\ & 3x_1^2 x_3 \ln \eta_{13} + 3x_2^2 x_3 \ln \eta_{23} + 3x_2 x_3^2 \ln \eta_{32} + 3x_1 x_3^2 \ln \eta_{31} + 3x_2^2 x_1 \ln \eta_{21} + 6x_1 x_2 x_3 \ln \eta_{123} \end{aligned} \quad (2)$$

$$M_{12} = (2M_1 + M_2)/3$$

$$M_{21} = (2M_2 + M_1)/3$$

$$M_{13} = (2M_1 + M_3)/3$$

$$M_{31} = (2M_3 + M_1)/3$$

$$M_{23} = (2M_2 + M_3)/3$$

$$M_{32} = (2M_3 + M_2)/3$$

$$M_{123} = (M_1 + M_2 + M_3)/3$$

$$M_{avg} = x_1 M_1 + x_2 M_2 + x_3 M_3 \quad (2a)$$

temperature were tested. Equations used to predict the viscosity of binary mixtures, were extended for ternary mixtures.

### Correlation with composition

The equations used for viscosity estimation of the ternary mixture are:

a) Grunberg- Nissan [17] equation:

$$\ln \eta_m = \sum_{i=1}^n x_i \ln \eta_i + \sum_{i=1}^n \sum_{j=i+1}^n x_i x_j G_{ij} \quad (1)$$

where  $\eta_m$  represents the viscosity of the mixture,  $\eta_i$  and  $x_i$  represents the viscosity and the molar fraction, respectively of component i of the mixture,  $G_{ij}$  is an interaction parameter, n represents the number of pure components in the mixture.

This equation was tested without interaction parameter, with three binary parameters ( $G_{ij}$ ), and with three binary and one ternary parameter ( $G_{123}$ ); the mass fraction ( $w$ ) was used instead of molar fraction to express mixture composition:

where  $M_i$  represents the molar mass of component  $i$ ,  $M_{ij}$  and  $\eta_j$  represent the binary parameters,  $M_{123}$  and  $\eta_{123}$  represent the ternary parameter.

c) Orbey and Sandler equation applied for petroleum products and extended for mixtures with biofuels [19]:

$$\eta = (w_1\eta_1^{1/3} + w_2\eta_2^{1/3} + w_3\eta_3^{1/3})^3 \quad (3)$$

d) An equation used in chemical engineering:

$$\eta = \frac{(x_1M_1) + (x_2M_2) + (x_3M_3)}{\frac{x_1M_1}{\eta_1} + \frac{x_2M_2}{\eta_2} + \frac{x_3M_3}{\eta_3}} \quad (4)$$

Using Statistic V10 software, an empirical polynomial equation was obtained:

$$\eta = aw_1 + bw_2 + cw_3 + dw_1w_2 + ew_1w_3 + fw_2w_3 \quad (5)$$

#### Correlation with temperature

The equations used to correlate the viscosity of ternary mixture containing isopropanol with temperature are the equations (6) and (7): Andrade equation and that used by Tat and Van Gerpen, respectively [20]:

$$\eta = a \cdot e^{b/T} \quad (6)$$

$$\ln \eta = a + \frac{b}{T} + \frac{c}{T^2} \quad (7)$$

where  $a$ ,  $b$  and  $c$  are correlation parameters,  $T$  is absolute temperature.

The accuracy of the equations used to calculate the viscosity of the ternary system was evaluated with the average relative deviation,  $ARD$  (%):

$$ARD = \frac{100}{n} \sum_i \frac{|\eta_{exp,i} - \eta_{cal,i}|}{\eta_{exp,i}} \quad (8)$$

where  $n$  is the number of experimental data points,  $\eta_{exp,i}$  and  $\eta_{cal,i}$  represent the experimental and calculated viscosity, respectively.

## Results and discussions

### The obtained experimental data

Experimental results of the viscosity of the ternary system biodiesel+diesel fuel+isopropanol over the entire composition range and for temperature ranging from 293.15 K to 323.15 K are presented in table 3. To better illustrate the influence of composition and temperature on the viscosity of the studied system, figure 1 presents viscosity variation with composition at 293.15 K and figure 2 presents viscosity variation with temperature. Viscosity variation with composition was similar at all the other investigated temperatures.

It was observed (table 3 and fig.1) that the dynamic viscosity of the ternary biodiesel (1)+diesel fuel (2)+isopropanol (3) system presents a variation with a minimum for each isothermal surface. Most viscosity values are within the values of the pure components, biodiesel, diesel fuel and isopropanol, respectively, but there are lower values than these. The viscosity of the ternary system decreases non-linearly with temperature increasing (fig.2).

### Viscosity calculation

The dynamic viscosity of the ternary system biodiesel+diesel fuel+isopropanol was calculated at different temperatures using eqs. (1-5). Several correlative equations: semiempirical Grunberg-Nissan equation (1a, 1c) and Mc Allister equation (2), an empirical equation (5) and predictive equations (eqs. 1a, 3, 4) have been used to estimate the viscosity of liquid mixtures from pure component data or to correlate the experimental data. The values of the correlative equations parameters were obtained by fitting the experimental data. The correlation parameters are shown in table 4 and the average relative deviation ( $ARD$ ) is presented in figure 3 for equations (1-4). The corresponding  $ARD$  values in viscosity calculation using eq. (5) are between 1 and 4 % for the studied domain of temperature. It can be observed that the greatest accuracy of viscosity estimation was obtained with Mc Allister equation (eq.2) ( $ARD$  under 0.03 %).

**Table 3**  
DYNAMIC VISCOSITY (mPa . s) OF THE TERNARY SYSTEM  
BIODIESEL (1)+DIESEL FUEL (2)+ISOPROPANOL (3) AT DIFFERENT TEMPERATURES

w1	w2	Temperature (K)						
		293.15	298.15	303.15	308.15	313.15	318.15	323.15
0.0966	0.0918	4.6687	4.1407	3.6633	3.2961	2.9487	2.6564	2.4032
0.2082	0.0990	4.4212	3.9208	3.4689	3.0889	2.7683	2.5078	2.2653
0.1960	0.1864	3.7992	3.3740	2.9843	2.6588	2.3820	2.1442	1.9402
0.0985	0.2809	3.5119	3.1146	2.7517	2.4462	2.1867	1.9894	1.7669
0.3921	0.0932	3.9072	3.4608	3.0650	2.7330	2.4514	2.2098	2.0041
0.3959	0.1882	3.3884	3.0025	2.6592	2.3698	2.1319	1.9164	1.7342
0.2984	0.2837	3.1924	2.8194	2.4928	2.2256	1.9919	1.7893	1.6152
0.1999	0.3802	3.0296	2.6747	2.3582	2.0915	1.8639	1.6662	1.4889
0.1005	0.4775	2.9208	2.5709	2.2631	2.0027	1.7805	1.5883	1.4281
0.1025	0.6822	2.5403	2.2223	1.9435	1.7078	1.5072	1.3339	1.1971
0.2040	0.5818	2.5457	2.2391	1.9655	1.7342	1.5360	1.3652	1.2175
0.4039	0.3840	2.7371	2.4053	2.1171	1.8744	1.6680	1.4916	1.3478
0.5999	0.1901	3.0161	2.6703	2.3766	2.1081	1.8893	1.7010	1.5393
0.5940	0.0941	3.5337	3.1294	2.7748	2.4771	2.2240	2.0081	1.8324
0.7999	0.0951	3.0961	2.7513	2.4405	2.2011	1.9689	1.7806	1.6088
0.6059	0.2880	2.7294	2.4127	2.1292	1.8908	1.6874	1.5094	1.3715
0.4080	0.4849	2.6154	2.3027	2.0220	1.7846	1.5744	1.4022	1.2516
0.3075	0.5848	2.5181	2.2062	1.9380	1.7159	1.5082	1.3378	1.1979
0.1036	0.7877	2.4426	2.1341	1.8562	1.6261	1.4341	1.2649	1.1201
1	0	6.1425	5.4376	4.7994	4.2660	3.8176	3.4366	3.1108
0	1	3.4249	3.0658	2.7330	2.4370	2.2032	1.9894	1.8132
0	0	2.3934	2.0628	1.7763	1.5373	1.3346	1.1684	1.0231

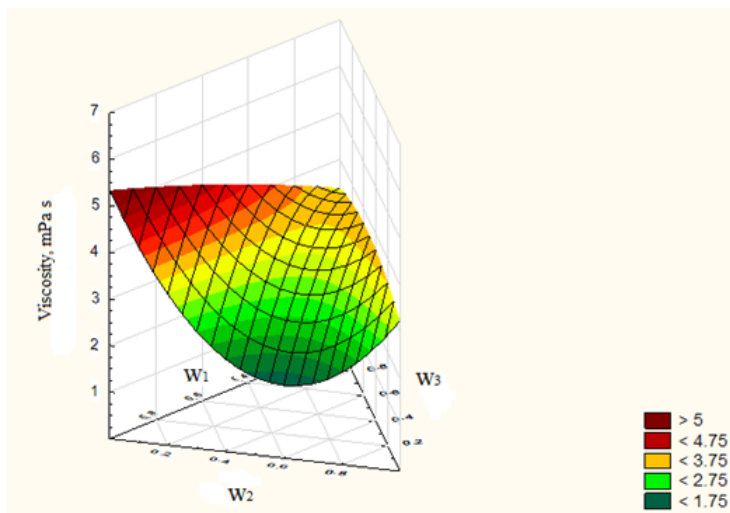


Fig.1. Viscosity variation with composition at 298.15 K for the ternary system biodiesel (1)+diesel fuel (2)+isopropanol (3)

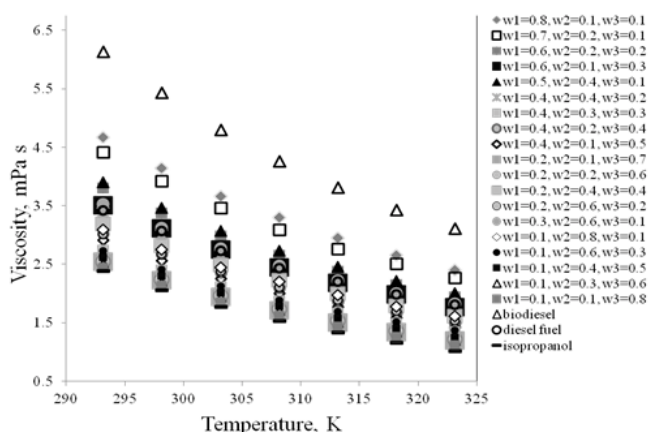


Fig. 2. Dynamic viscosity variation with temperature for the ternary biodiesel(1)+diesel fuel(2)+isopropanol(3) system at different compositions

**Table 4**  
VISCOSITY CORRELATION PARAMETERS WITH COMPOSITION FOR THE TERNARY SYSTEM BIODIESEL (1) + DIESEL FUEL (2) + ISOPROPANOL (3) AT DIFFERENT TEMPERATURES

Equation	Param.	Temperature (K)						
		293.15	298.15	303.15	308.15	313.15	318.15	323.15
Grunberg-Nissan (1b)	G <sub>12</sub>	-3.8561	-3.9536	-4.0303	-4.1044	-4.2286	-4.3386	-4.4142
	G <sub>13</sub>	0.9587	1.0723	1.1938	1.3183	1.4613	1.5904	1.7486
	G <sub>23</sub>	1.0276	1.0370	1.0407	1.0647	1.0757	1.0988	1.0361
Grunberg-Nissan (1c)	G <sub>12</sub>	-3.9440	-3.9536	-4.0303	-4.1044	-4.2286	-4.4680	-4.5565
	G <sub>13</sub>	0.9042	1.0723	1.1938	1.3183	1.4613	1.4675	1.6135
	G <sub>23</sub>	0.9805	1.0370	1.0407	1.0647	1.0757	0.9723	0.8970
	G <sub>123</sub>	0.9244	1.0723	1.1938	1.3183	1.4613	1.5904	1.7486
McAllister (2)	$\eta_{12}$	3.8021	3.5636	3.2929	3.0974	2.9262	2.7867	2.4873
	$\eta_{13}$	0.8082	0.6104	0.4743	0.4066	0.2918	0.2368	0.1951
	$\eta_{23}$	1.6773	1.4126	1.1916	1.0242	0.8664	0.7534	0.5996
	$\eta_{21}$	1.6843	1.4109	1.1951	1.0316	0.8574	0.7294	0.6481
	$\eta_{31}$	0.6342	0.4685	0.3442	0.2631	0.1873	0.1426	0.0914
	$\eta_{32}$	24.8231	23.5806	22.1220	21.4790	20.3555	19.6469	19.9573
	$\eta_{123}$	39.6830	41.6233	43.7186	42.0893	48.7200	50.4523	60.1433
Polynomial (5)	a	5.3396	4.7306	4.1793	3.7231	3.3323	3.0034	2.7179
	b	3.2338	2.8762	2.5481	2.2611	2.0323	1.8232	1.6556
	c	3.3898	2.9671	2.5892	2.2845	2.0134	1.7937	1.5957
	d	-10.6529	-9.6061	-8.5992	-7.7449	-7.0933	-6.5067	-5.9659
	e	0.2037	0.3591	0.5116	0.5941	0.7172	0.7653	0.8429
	f	1.3773	1.2097	1.0551	0.9417	0.8333	0.7680	0.6269

It must be mentioned that this equation can be applied only if the mean molar masses of biodiesel and diesel fuel are available. Also, a good accuracy was obtained when using Grunberg and Nissan equation with three parameters (binary interaction parameters) (eq.1b) and with four

parameters (binary and ternary interaction parameters) (eq.1c) (*ARD* under 0.06 %). The *ARD* values decrease with temperature increasing. Taking into account that Grunberg and Nissan equation with four parameters (eq.1c) provides

**Table 5**  
REGRESSION PARAMETERS AND ERRORS FOR DYNAMIC VISCOSITY CORRELATION WITH TEMPERATURE AT VARIOUS COMPOSITIONS OF THE PSEUDO-TERNARY SYSTEM

w1	w2	Eq. (6)			Eq. (7)			
		Parameter		ARD (%)	Parameter			ARD (%)
		a	b		a	b	c	
0.0966	0.0918	-5.6141	2097.00	0.0576	-4.3598	1324.9	118691	0.0406
0.2082	0.0990	-5.7366	2116.80	0.1076	-2.5881	178.7	297943	0.0575
0.1960	0.1864	-5.9371	2131.70	0.0622	-4.2433	1089.0	160291	0.0419
0.0985	0.2809	-6.1045	2157.80	0.1153	-5.6074	1851.0	47041	0.1145
0.3921	0.0932	-5.8555	2115.30	0.0842	-2.9196	308.1	277828	0.0317
0.3959	0.1882	-6.0080	2118.50	0.0602	-3.9565	855.7	194136	0.0344
0.2984	0.2837	-6.1814	2151.70	0.0482	-4.4230	1069.3	166398	0.0189
0.1999	0.3802	-6.5400	2242.70	0.0374	-7.5393	2857.9	-94571	0.0314
0.1005	0.4775	-6.6641	2267.90	0.0397	-5.8709	1779.6	75066	0.0343
0.1025	0.6822	-7.2253	2391.60	0.0643	-6.2698	1803.4	90422	0.0645
0.2040	0.5818	-7.0266	2334.60	0.0419	-8.3754	3165.0	-127646	0.0263
0.4039	0.3840	-6.6637	2248.00	0.0904	-3.7219	437.1	278890	0.1678
0.5999	0.1901	-6.1710	2132.50	0.0613	-4.7046	1229.8	138769	0.0511
0.5940	0.0941	-5.8558	2085.40	0.1274	-1.3448	-691.4	426882	0.0484
0.7999	0.0951	-5.9128	2064.30	0.0586	-5.2137	1634.0	66161	0.0538
0.6059	0.2880	-6.4703	2190.80	0.0925	-4.0033	672.2	233456	0.0704
0.4080	0.4849	-7.0088	2337.60	0.0620	-8.4388	3217.8	-135323	0.0490
0.3075	0.5848	-7.1101	2355.70	0.0664	-8.3877	3142.2	-120908	0.0596
0.1036	0.7877	-7.5163	2466.10	0.0458	-8.7584	3230.7	-117544	0.0367
1	0	-5.5507	2158.80	0.0927	-2.5509	312.3	283870	0.0481
0	1	-5.6708	2023.50	0.0820	-4.1530	1089.3	143629	0.0784
0	0	-8.2973	2689.10	0.0481	-9.4921	3424.6	-113067	0.0334

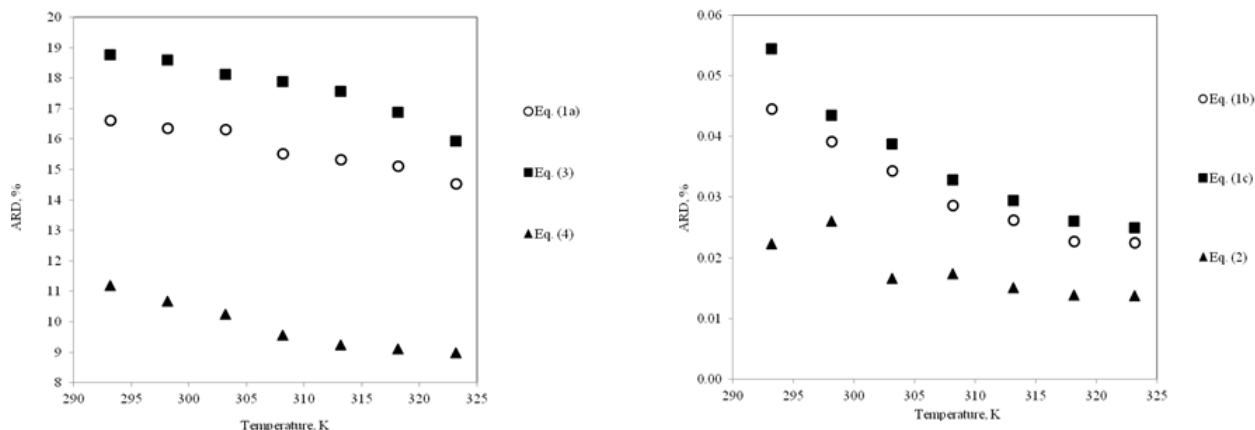


Fig.3. Average relative deviation (ARD, %) values for viscosity calculation with a) Eqs. (1a, 3, 4); b) Eqs. (1b, 1c, 2)

only a slightly accuracy increasing, eq. (1b) can be recommended for the ternary studied system viscosity estimation.

All predictive equations: Grunberg and Nissan equation without interaction parameter (eq. 1a), and eqs.(3) and (4) give relatively great errors in viscosity calculation and they cannot be recommended for this ternary system with isopropanol.

Regarding the viscosity variation with temperature, the data are well represented by the classical Andrade and Tat & Van Gerpen equations. Table 5 presents the values of correlation parameters from equations (6,7) and the corresponding errors (ARD, %) for the ternary system biodiesel+diesel fuel+isopropanol viscosity estimation in the temperature range of 293.15 K to 323.15 K. The two tested equations (6) and (7) have similar accuracies (ARD under 0.1 %), both of them being recommended for viscosity estimation at different temperatures for the ternary studied system.

### Conclusions

Experimental data of the viscosity of the pseudo-ternary biodiesel+diesel fuel+isopropanol system over the entire

composition range and for temperature ranging from 293.15 K to 323.15 K were reported. A variation with minimum values of the viscosity with composition was registered and a variation with temperature following the classical Andrade and Tat & Van Gerpen equations was obtained. The viscosity of the ternary system can be accurately estimated in the investigated temperature range (ARD under 0.06 %) using Grunberg and Nissan equation with three parameters or Mc Allister equation with seven parameters. The viscosity can be accurately calculated at different temperatures in the range of 273.15 K to 323.15 K using exponential equations.

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Manuscript received: 1.11.2017