

## Dynamic viscosity dependence on temperature for fuels used for diesel engine

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**Abstract.** Viscosity is an important property of fuels used for diesel engine affecting engine's efficiency and harmful gases emission. Viscosity of liquid fuels depends especially on fuels composition and temperature. The dynamic viscosity of diesel fuel, biodiesel and blends of diesel with biodiesel, *i*-propanol and *n*-butanol was measured for temperature ranging from 293.15 K to 323.15 K and atmospheric pressure. It has been verified that well-known Arrhenius derived equations can be used to estimate with good accuracy, viscosity at different temperatures for diesel, biodiesel, diesel+biodiesel blends, but also for diesel blends with propanol and butanol. Values of activation parameters: activation energy, activation enthalpy and activation entropy for the viscous flow were derived based on linearized Eyring's type equation. The values of the activation energy for viscous flow of fuels and fuels blends calculated based on measured values of dynamic viscosity in the temperature range of 273.15 K and 323.15 K were similar to those presented in the literature for some hydrocarbons, esters, and alcohols, respectively.

**Keywords:** dynamic viscosity; diesel, biodiesel; activation energy; viscous flow.

### 1. Introduction

Due to the concern raised by the increased environment pollution and the need to preserve oil resources, a trend has emerged in the last years, concerning fuels used in transportation: the use of biofuels as additives and/or substitutes for fossil fuels. Today, diesel fuel is commercialized blended with biodiesel. Other biofuels like bioalcohols are considered to be promising alternatives to diesel fuel.

Viscosity is an important property of fuels used for diesel engine [1, 2]; it is related to fluid flow process and affects the fuel injection into the combustion chamber and the atomization quality [3-5], influencing fuel combustion and affecting engine's efficiency and harmful gases emission [6, 7].

Viscosity of liquid fuels depends especially on fuels composition and temperature, the pressure having a much lower influence. The dependency of the viscosity of diesel, biodiesel, and their blends on temperature was extensively investigated in the literature in the recent years [6, 8-14]. A reduced number of studies have addressed the issue of viscosity for diesel and biodiesel with bioalcohols blends [2, 15-18]. Diesel fuel and biodiesel follow the general behavior of liquids, their viscosity decreasing non-linearly with temperature increasing [1, 13, 14, 19, 20, 21]. Modelling the viscosity dependence on temperature for diesel fuel and its blends with biofuels was often done with an Arrhenius type equation [5, 13].

Based on Arrhenius equation, the energy of activation for the viscous flow can be derived. Values of energy of activation for the viscous flow for pure

substances like hydrocarbons, alcohols, esters and their mixtures are reported in the literature [21-24].

The Gibbs free energy of activation for viscous flow derived from viscosity data for methylcyclohexane has been reported of 13.3 kJ/mol (298.15 K) and varies for some methyl esters of fatty acids from 17.19 kJ/mol (methyl decanoate) to 19.37 kJ/mol (methyl tetradecanoate) [24]. Greater values of free energy of activation for viscous flow were reported in the literature at 298.15 K for methanol (43.427 kJ/mol), ethanol (46.426 kJ/mol), and *n*-propanol (49.024 kJ/mol) [23].

The activation enthalpy for viscous flow for hydrocarbons like heptane and octane, alcohols like 1-pentanol and 1-hexanol and their mixtures were calculated from the intercept of the straight lines representing the Gibbs free energy of activation for viscous flow dependence on temperature [21]. The activation enthalpy for the viscous flow for pure *n*-pentanol and *n*-hexanol was found to be approximately 2.6 times greater than the activation enthalpy for the viscous flow for *n*-heptane and *n*-octane, respectively. The activation enthalpy for the viscous flow at 298.15 K of some methyl esters of fatty acids (methyl decanoate, methyl dodecanoate, methyl tetradecanoate) were found to be of approximately 1.5 to 1.9 times greater than that of a hydrocarbon like methylcyclohexane [24].

As far as we know, values of energy of activation for the flow process for multicomponent systems like diesel, biodiesel and their blends with bioalcohols are not reported in the literature.

The aim of this paper is the study of dynamic viscosity dependence on temperature for diesel fuel, biodiesel, diesel+biodiesel, diesel+*i*-propanol, and

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diesel+*n*-butanol blends in terms of energy of activation for the viscous flow. The dynamic viscosity of diesel fuel, biodiesel and blends of diesel with biodiesel, *i*-propanol and *n*-butanol was measured for temperature ranging from 293.15 K to 323.15 K and atmospheric pressure. It has been verified that models derived from the well-known Arrhenius equation can be used to estimate with good accuracy, viscosity at different temperatures for diesel, biodiesel, diesel+biodiesel blends, but also for diesel blends with propanol and butanol. The dynamic viscosity dependence on temperature of diesel fuel, biodiesel and diesel blends with biodiesel, *i*-propanol and *n*-butanol respectively, is studied using Eyring's reaction rate theory and activation parameters for viscous flow are derived. Activation energy plays an important role in the evaluation of liquid flow properties. The values of activation energy for viscous flow could be used for analysis of molecular interactions existing in the blends.

## 2. Experimental

Diesel fuel and biodiesel were procured from a local company and they fulfil European standards quality requirements, EN 590 for diesel fuel and EN 14214 for biodiesel, respectively. *i*-Propanol of 99.0 % purity, and *n*-butanol of 99.5 % purity were supplied by Chemical Company, Romania. Properties of these fuels are listed

**Table 1.** Properties of diesel fuel, biodiesel, *i*-propanol and *n*-butanol

Property	Diesel	Biodiesel	<i>i</i> -Propanol	<i>n</i> -Butanol
Density at 20 °C, g/cm <sup>3</sup>	0.8390	0.8846	0.7859	0.8114
Kinematic viscosity at 40 °C, mm <sup>2</sup> /s	3.0110	4.5620	1.7438	2.2622
Cetane number	46	52	12*	16*

\*from literature [25]

### 2.1. Equations for viscous flow of liquid fuels

An Arrhenius type equation was often used to express the viscosity dependence on temperature for diesel, biodiesel and their blends [5, 12]:

$$\eta = A \exp\left(\frac{E_a}{RT}\right) \quad (1)$$

where  $\eta$  represents the dynamic viscosity;  $E_a$  - activation energy for viscous flow;  $R$  - ideal gas constant;  $T$  - absolute temperature;  $A$  - pre-exponential constant.

Based on Eyring's reaction rate theory, the Gibbs free energy of activation for the flow process can be calculated:

$$\eta = \frac{hN_A}{V} \exp\left(\frac{\Delta G^\ddagger}{RT}\right) \quad (2)$$

where  $h$  is Planck's constant;  $N_A$  - Avogadro's number;  $V$  - molar volume;  $\Delta G^\ddagger$  - free energy of activation for viscous flow.

Equation (2) can be linearized by applying the logarithm function:

$$\ln \eta = \ln \frac{hN_A}{V} + \frac{\Delta G^\ddagger}{R} \frac{1}{T} \quad (3)$$

The enthalpy and entropy of activation for viscous flow can be obtained if in Eq. (3), the free energy of activation for viscous flow is made explicit according to its thermodynamic definition relationship:

in Table 1. Binary blends of diesel with biofuels were prepared by using an analytical balance having an accuracy of 0.0001 g. Diesel+biodiesel blends with mass fraction of biodiesel of 0.105; 0.260; 0.513, and 0.760 were prepared. Diesel+biodiesel blends were named Bdx.xxx, where x.xxx represents the mass fraction of biodiesel in the blend. Alcohols blends with diesel fuel were named Prox.xxx and Bux.xxx for diesel+*i*-propanol and diesel+*n*-butanol blend, respectively. Diesel+*i*-propanol blend with 0.094 mass fraction of alcohol was named Pro0.094, and diesel+*n*-butanol blend with 0.097 mass fraction of butanol was named Bu0.097.

Dynamic viscosity was measured at atmospheric pressure by using an Anton Paar Stabinger viscometer model SVM 3000. The device has been previously described [12, 21]. It is equipped with two measuring cells and provides the dynamic viscosity and density of the fuel sample at a desired temperature. The viscometer has a built-in Peltier thermostat which allows maintaining the sample temperature with an accuracy of  $\pm 0.01$  K. The uncertainty of the dynamic viscosity is 0.35 % and the uncertainty for density is 0.0005 g/cm<sup>3</sup>. The measurements were done between 293.15 K and 323.15 K. Double-distilled water and dry air were used as reference fluids for the viscometer calibration.

$$\ln\left(\frac{\eta}{\frac{hN_A}{V}}\right) = \frac{\Delta H^\ddagger}{R} \times \frac{1}{T} - \frac{\Delta S^\ddagger}{R} \quad (4)$$

where  $\Delta H^\ddagger$  and  $\Delta S^\ddagger$  are the enthalpy and the entropy of activation for viscous flow, respectively.

## 3. Results and discussion

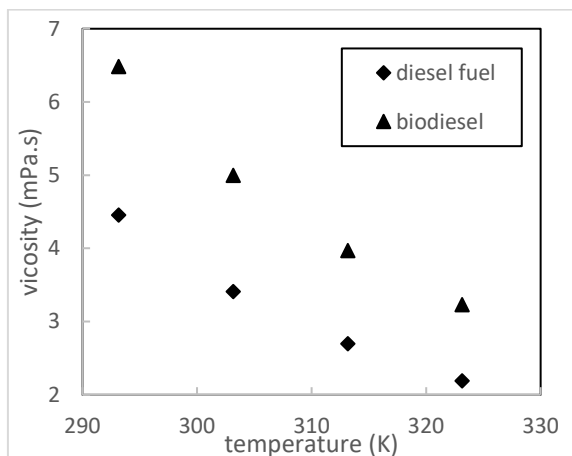
The measured dynamic viscosity variation with temperature for diesel fuel and biodiesel (Fig. 1), diesel+biodiesel blends (Fig. 2), and diesel+*i*-propanol and diesel+*n*-butanol (Fig. 3) is graphically presented in order to better highlights the influence of temperature on the viscosity of these fuels. A nonlinear decrease in viscosity with temperature increasing for all investigated fuels and fuels blends as expected, was observed (Fig. 1 –3). The influence of temperature on viscosity is more pronounced at low temperatures. A sharp initial decrease in viscosity with temperature increasing was registered for all investigated fuels and blends.

Empirical equations derived from the best known Arrhenius equation were used to calculate the viscosity of the studied fuels blends:

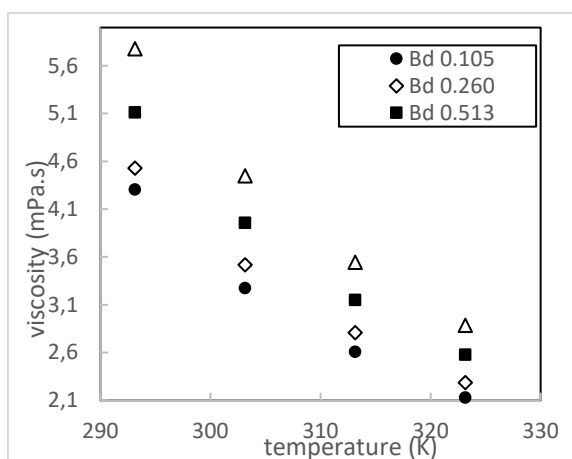
$$\eta = \exp\left(A + \frac{B}{C+T}\right) \quad (5)$$

$$\eta = \exp\left(A + \frac{B}{T} + \frac{C}{T^2}\right) \quad (6)$$

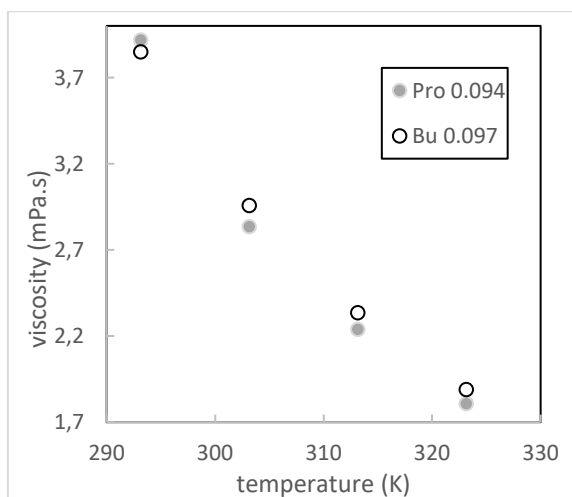
where A, B, C are regression parameters.



**Figure 1.** Viscosity versus temperature for diesel fuel and biodiesel



**Figure 2.** Viscosity versus temperature for diesel+biodiesel blends



**Figure 3.** Viscosity versus temperature for diesel+i-propanol and diesel+n-butanol blends

The accuracy of viscosity estimation was evaluated in terms of average absolute deviation (AAD %) calculated by the equation:

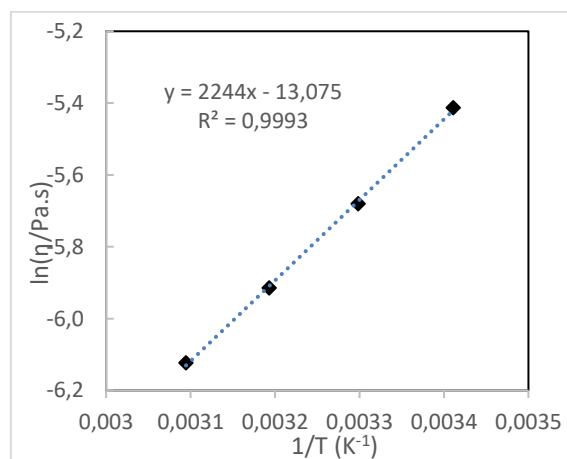
$$AAD = \frac{100}{N} \sum_{i=1}^N \frac{|\eta_{exp,i} - \eta_{cal,i}|}{\eta_{exp,i}} \quad (7)$$

where N is the number of experimental points;  $\eta_{exp}$ ,  $\eta_{cal}$  – experimental and calculated values of viscosity.

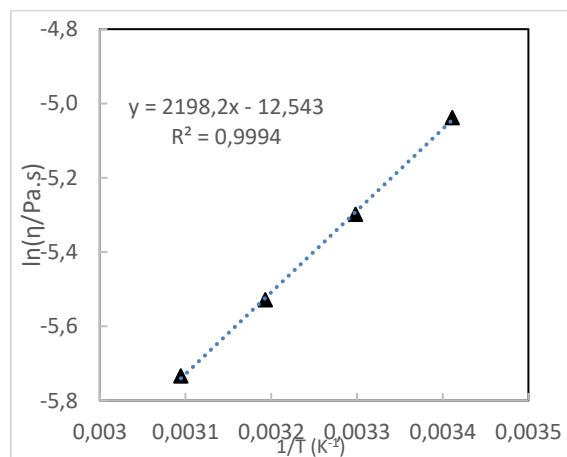
Table 2 presents the value of regression parameters from Eqs. (5) and (6) and the corresponding AADs for the calculated dynamic viscosity of diesel+biodiesel blends and diesel+i-propanol and diesel+n-butanol blends for temperature ranging from 293.15 K to 313.15 K. It can be seen (Table 2) that Equation (5) estimates with good accuracy (AAD lower than 1.85%) the viscosity dependence on temperature for the studied blends in the temperature range of 293.15 K to 323.15 K while the accuracy of Equation (6) is lower (AAD ranged from 13.23 % to 16.31 %).

Straight lines were obtained by plotting  $\ln(\eta)$  versus  $1/T$  (linearized Arrhenius equation, Eq.1) for all investigated fuels and fuels blends, the correlation coefficient varying between 0.9950 and 0.9997.

The graphical representations obtained for diesel and biodiesel are presented as example (Fig. 4). Derived values for activation energy of viscous flow,  $E_a$ , pre-exponential constant A, and the value of coefficient of correlation  $R^2$  are presented in Table 3.



a



b

**Figure 4.** Graphical representation of linearized Eq. (1) for diesel fuel (a: ♦) and biodiesel (b: ▲). Experimental data (points) and correlation data ('---')

**Table 2.** Values of regression constants A, B, and C from Equations (5) and (6) and corresponding AAD (%) for the studied fuels and fuels blends

Fuel	A	B	C	AAD (%)
Eq. (5)				
Bd0.105	-1.94	392.82	-177.80	0.14
Bd0.260	-5.63	2017.69	-10.29	0.40
Bd0.513	-5.51	2008.92	-11.47	0.63
Bd0.760	-5.41	1999.41	-13.83	0.61
Pro0.094	-6.50	2165.37	-17.21	1.86
Bu0.097	-6.07	2089.60	-11.09	0.50
Eq. (6)				
Bd0.105	1.46	1.18	0.95	13.23
Bd0.260	1.51	1.25	1.03	14.04
Bd0.513	1.63	1.37	1.14	15.27
Bd0.760	1.75	1.49	1.26	16.31
Pro0.094	1.36	1.04	0.80	11.14
Bu0.097	1.34	1.08	0.84	11.75

**Table 3.** Activation energy for viscous flow  $E_a$ , constant A, and coefficient of correlation  $R^2$  from linearized form of Eq. (1)

Fuel	$E_a$ (kJ/mol)	A (mPas)	$R^2$
Diesel	18.657	0.0021	0.9993
Biodiesel	18.276	0.0036	0.9994
Bd0.105	18.458	0.0022	0.9984
Bd0.260	17.950	0.0029	0.9997
Bd0.513	18.013	0.0031	0.9993
Bd0.760	18.220	0.0033	0.9994
Pro0.094	20.191	0.0010	0.9950
Bu0.097	18.689	0.0018	0.9996

Values of the activation energy for viscous flow close to those presented in Table 3 for diesel fuel (18.657 kJ/mol) were found in the literature for benzene (14.955 kJ/mol, [26]) and *n*-heptane (14.955 kJ/mol, [27]). The value of the activation energy found in the literature for *i*-propanol (25.430 kJ/mol [28] and 21.950 kJ/mol [29]) was close to that presented in Table 3 for diesel+*i*-propanol blend, with low content of alcohol (Pro0.094: 20.191 kJ/mol).

As molecular motion, the flowing process of liquids depends on the interactive force among molecules so, it is related to the energy of activation for viscous flow. The energy of activation represents the minimum energy required for a molecule during the flow process to overcome the internal energy barrier, and to move from the original position to a nearby one represented by a hole. The energy of activation corresponds to this energetic state and the pre-exponential constant is correlated to the disorder corresponding to the viscous flow, so it is an entropic nature factor. It can be observed (Table 3) that the activation energy for viscous flow has relatively close values for diesel, biodiesel and their blends, ranging between 17.95 kJ/mol and 18.65 kJ/mol. The activation energy for viscous flow for blends of diesel with low content of biodiesel, *i*-propanol and *n*-butanol respectively, varied in the order: 18.458 kJ/mol (Bd0.105) < 18.689 kJ/mol (Bu0.097) < 20.191 kJ/mol (Pro0.094).

The values of activation parameters for the viscous flow,  $\Delta H^\ddagger$  and  $\Delta S^\ddagger$  for the studied fuels and fuels blends were obtained from the graphical representation of  $\ln(\eta) - \ln(hN_A/V)$  versus  $1/T$ . In this sense, for the constants grouping  $(hN_A/V)$ , the value of the constant A calculated based on the graphical representation of Eq. (1) has been considered. Straight lines were obtained for all studied

fuels and fuels blends by graphically representing Eq. (4). The entropy of activation for viscous flow was derived from the intercept with the ordinate, and the enthalpy of activation for viscous flow was calculated from the slope of the straight line. The graphical representations obtained for Bd0.105, Pro0.094 and Bu0.097 blends are presented as example in Fig. 5.

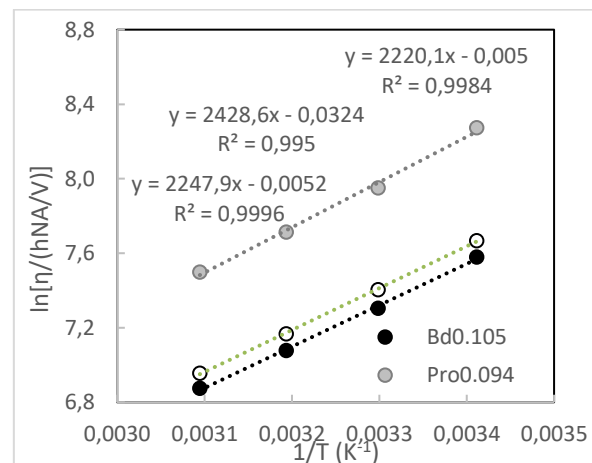
**Figure 5.** Graphical representation of Eq. (4) for calculation of activation parameters for the viscous flow. Experimental data (points) and correlation data ('---')

Table 4 presents the values of the enthalpy ( $\Delta H^\ddagger$ ) and entropy ( $\Delta S^\ddagger$ ) of activation for viscous flow respectively, for the investigated fuels and fuels blends. Close values of the enthalpy of activation for viscous flow have been obtained for the studied fuels and fuels blends. The greatest value of  $\Delta H^\ddagger$  (20.191 kJ/mol) corresponds to diesel+*i*-propanol blend (Pro0.094), and the smallest value (17.950 kJ/mol) corresponds to

diesel+biodiesel blend (Bd0.260). The values of enthalpy of activation for viscous flow obtained for diesel, biodiesel and their blends are similar with values reported in the literature for methylcyclohexane, a diesel surrogate, and its blends with three different fatty acid methyl esters (methyl decanoate, methyl dodecanoate and methyl tetradecanoate) [24]. Values of  $\Delta H^\ddagger$  very close to those obtained in this study for blends of diesel with *i*-propanol and *n*-butanol have been reported in the literature for mixtures of hydrocarbons (heptane or octane) with alcohols (*n*-pentanol or *n*-hexanol) [21].

The contribution of the entropy of activation to the free energy of activation for the viscous flow is small (Table 4), resulting that the process is enthalpy controlled for the investigated fuels

**Table 4.** Values of the enthalpy ( $\Delta H^\ddagger$ ) and entropy ( $\Delta S^\ddagger$ ) of activation for viscous flow

Fuel	$\Delta H^\ddagger$ (kJ/mol)	$\Delta S^\ddagger$ (J/mol·K)
Diesel	18.657	0.0100
Biodiesel	18.276	0.0690
Bd0.150	18.458	0.0416
Bd0.260	17.950	0.1305
Bd0.513	18.013	-0.0914
Bd0.760	18.220	0.1189
Pro0.094	20.191	0.2694
Bu0.097	18.689	0.0432

#### 4. Conclusions

Experimental data of dynamic viscosity for diesel, biodiesel, diesel+biodiesel blends, and diesel+*i*-propanol, and diesel+*n*-butanol blends with low alcohol content are reported for temperature ranging from 293.15 K to 323.15 K. Empirical equations derived from Arrhenius equation were tested to estimate dynamic viscosity of studied fuels at different temperatures. Eq. (5) gives good results (AAD < 1.86 %) in estimating the viscosity at different temperatures for all the studied systems.

It was observed that the studied fuels and fuels blends in the temperature range of 273.15 K to 323.15 K obey to the exponential Arrhenius behavior. Based on experimental dynamic viscosity data, values of two viscosity Arrhenius parameters, the activation energy for viscous flow and the pre-exponential constant were determined and compared with literature data.

The values calculated for the activation parameters of the viscous flow,  $\Delta H^\ddagger$  and  $\Delta S^\ddagger$  show that the flow process is enthalpy controlled for all the studied fuel systems.

#### Conflict of interest

Authors declare no conflict of interest.

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