

Pour Point and Predictive Models for the Viscosity-Temperature Non-Linear Behaviour of Ternary Fuel Blends for a Compression Ignition Engine

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Due to the ever-increasing consumption of fossil fuels, their impact on the environment, and the volatility of the market, it is advisable to use biofuels that can be produced locally from renewable sources, which supports the local economy, agriculture, and related processing industries. This article deals with how to improve the flow properties and pour point of biofuels for diesel engines. For the experiment, biodiesels in the form of rapeseed methyl ester (RME) and methyl ester made from waste animal fats and tallow (WAFME) were used. The pour point, viscosity, and density of WAFME were improved by mixing with RME and/or by adding bio-alcohols (alcohols produced from biomass, e.g., lignocellulosic). All used biofuels were classified as 2nd generation biofuels. The flow properties of the mixtures were monitored and subsequently modelled at temperatures from -10 to 60 °C. The addition of bio-alcohol had a statistically significant effect on the decrease in the viscosity and pour point of ternary blends ($p < 0.05$). Mathematical models of the dependence of kinematic viscosity on the temperature of mixtures (power law, exponential, Arrhenius, and Vogel) were created.

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INTRODUCTION

The energy demands of the 21st century are challenging. Environmental degradation and climate change are a threat that the European Union (EU) and the world must face. Due to the uncertain situation in the oil market, which we are now observing, for example, as a result of the sanctions directed against the Russian Federation, it is appropriate to develop other local, ecological, and sustainable solutions (Arshad *et al.* 2018; Milessi *et al.* 2022). To solve the mentioned challenges, the 2030 EU climate & energy framework (European Commission 2017) was created, and, in December 2019, The European Green Deal (European Commission 2019) was announced. It determines the direction of the EU until 2050. The European Green Deal applies to all sectors of the economy, especially transport, energy, agriculture, *etc.* The possibility of meeting the set goals necessitates the greater use of biofuels, especially 2nd generation biofuels (Math *et al.* 2010). These are fuels for which the primary source, petroleum oil, is replaced by

another non-food input raw material, for example plant materials, animal waste, or used cooking oil (Sajjadi *et al.* 2016; Milessi *et al.* 2022).

Therefore, instead of using biodiesel fatty acid methyl ester (FAME), most often rapeseed methyl ester (RME), it is possible to use rendering and waste animal tallow and fats for the production of waste animal fatty methyl ester (WAFME) (Sirviö *et al.* 2019). The production of biodiesel from rendering and waste animal tallow and fats has great potential, as this feedstock leads to a global waste reduction and does not compete with the food industry (Sander *et al.* 2018). For an overview, it can be stated that the increasing population increases slaughterhouse wastes that create pollution (Bhunia *et al.* 2022; Mozhiarasi and Natarajan 2022). Each year, millions of animals are butchered around the world. Hernández-Fydrych *et al.* (2019) reported that cattle meat production in Mexico during the year 2008 to 2017 was 18,026,089 tons from 84,490,490 slaughtered animals. Singh *et al.* (2014) stated that from April 2009 to March 2013, nearly 8 million cattle were slaughtered in India. In the year of 2014, the total number of animals that were slaughtered during the pilgrimage season was about 2.5 million in Saudi Arabia (Nizami *et al.* 2017). Poland produces 380 tons of animal fat per year, and this can be used for transesterification purposes (Cisek 2018). Animal slaughter waste from goats as well as sheep is about 12% per unit of weight of the body, whereas, for cattle, it is around 38% per body weight (Nizami *et al.* 2017). Transesterification is an effective procedure generally used to convert animal fats and wastes to biodiesel (Arefin *et al.* 2021; Hasan and Ratnam 2022). Chakraborty *et al.* (2014) reported that slaughterhouse animal fat biodiesel properties were likely similar to that of traditional diesel fuel. For example, Chowdhury *et al.* (2022) stated that 4.785 million liters of chicken fat oil could generate around 4.068 million liters of biodiesel. However, the use of WAFME itself as an additive to diesel fuel is not possible due to the high amount of sulfur and its viscous properties, especially at temperatures below 10 °C (Golimowski *et al.* 2017). In order to achieve the obligation of adding 7% by volume of a biocomponent according to the Renewable Energy Directive (RED) (European Commission 2018), it is advisable to create a mixture of diesel, WAFME, and RME, which also achieves a more suitable viscosity and a lower sulfur concentration.

Another possible step to reduce the viscosity of the three-component mixtures for diesel engines is the addition of a small amount of alcohol (ethanol or butanol) (Wai *et al.* 2022). The viscosity is sharply reduced with a small content of any alcohol (Kumbár *et al.* 2015). This decreasing trend is not linear with respect to the volumetric, mass or molar content. The reduction is inversely proportional to the alcohol carbon chain, this trend being consistent with the viscosity of pure alcohols (Lapuerta *et al.* 2015). The addition of bio-alcohol will also increase the total amount of the bio-component. Another advantage of a bio-alcohol is a reduction in the amount of soot and an increase in the engine efficiency due to enriching the fuel with oxygen (Barrientos *et al.* 2013; Hemanth *et al.* 2022).

The viscosity and density are important properties of biodiesel that have a great influence on the performance characteristics of the engine. A critical variable is the size of the microdroplets injected into the piston chamber of the combustion engine (Hoang 2021; Younis *et al.* 2021). The size of the microdroplets also affects the fuel consumption and the quality of the combustion process (Razzaq *et al.* 2020). In the case of high viscosity, the filters may become clogged or the pumping of fuel can even be prevented (Refaat 2009). Barabas *et al.* (2010) notes that the diesel–biodiesel–ethanol blends (D85B10E5, D70B25E5, B80D10E10) have a very close density to diesel fuel on the whole considered temperature domain. The same author also claimed that the blend's viscosity is very close to that of diesel, and the differences get smaller with temperature increase. In general, both

RME and WAFME have a higher viscosity than diesel (Rakopoulos *et al.* 2007). The freezing point of pure WAFME is around 11 °C, which is mainly caused by the presence of saturated acids (Golimowski *et al.* 2017).

Siva *et al.* (2019) concluded that the performance of diesel engines powered by biodiesel blends was a little lower (overall average 42.97 kW) than that of diesel engines powered by diesel fuel (43.25 kW). According to Abed *et al.* (2018) and Siva *et al.* (2019), this phenomenon occurs because the calorific value of diesel fuel is higher than biodiesel. Reang *et al.* (2022) tested diesel–biodiesel–rice wine alcohol and state that at full load D80B10RW10, D70B20RW10, and D60B30RW10 gives 1.46%, 2.86%, and 3.91% lower brake thermal efficiency (BTHE) than diesel used in single cylinder compression ignition engine. Datta and Mandal (2017) found that the BTHE and fuel efficiency of a compression ignition engine increases while using alcohol (methanol and ethanol) with biodiesel. Nair *et al.* (2017) analyzed that the low percentage of biodiesel has high performance and lower emission than conventional fuel, which means that it can be easily introduced in the existing compression ignition engine without any modification. Mofijur *et al.* (2016) studied the ternary blends of ethanol–biodiesel–diesel and found that 5% to 10% ethanol share with 20% to 25% biodiesel share along with diesel reduced CO and HC emission. Hulwan and Joshi (2011) tested diesel–biodiesel–ethanol blend (D70B10E20). It was found that brake specific fuel consumption is increased but the brake thermal efficiency is improved for high ethanol content blends. The combustion process of a high ethanol content blend is delayed at low loads but approached to that of the diesel fuel at high loads. Cisek (2018) reported that hourly advanced motor fuel (AMF) consumption is about 9% higher than for standard diesel fuel (DF). The overall efficiency of the engine is lower by about 7% for the AMF-powered engine than for DF fueled engine. Use of AMF and DF decreases CO, HC, and smoke concentration (up to 37%), at the expense of increasing NOx in exhaust gases (about 4%) (Cisek 2018).

The aim of the present work was to determine and subsequently model the viscosity-temperature behaviour of ternary blends in the temperature range from -10 to 60 °C, which corresponds to the climatic conditions of most European countries.

The main hypothesis of the research is that by means of a suitable ratio of a mixture of diesel fuel, WAFME, RME, and bioethanol or biobutanol, very similar flow properties (including the pour point) to pure diesel fuel can be achieved, or alternatively diesel with the mandatory addition of biodiesel.

EXPERIMENTAL

Material

Fuel blend characterization

The fuel characterisation was carried out in the laboratories of the Faculty of AgriScience at Mendel University in Brno. The diesel without the biocomponent used in this study was supplied by Čepro (Střelice, Czech Republic), which follows European standard EN 590 (2013+A1:2017). The RME was donated by Preol (Lovosice, Czech Republic) and meets all the standards according to EN 14214 (2013+A2:2019). The WAFME was donated by Temperator (Liberec, Czech Republic), where Baynox (300 mg/kg) was used as the antioxidant. Two bio-alcohols were used – ethanol and butanol, both supplied by Lach-Ner (Neratovice, Czech Republic). The FAME and WAFME were

analysed within 1 week after production, and all the samples were stored at 20 °C in the dark, just like the samples in article (Sirviö *et al.* 2019).

For this study, mixtures with different contents of diesel fuel, RME, WAFME, bioethanol, or biobutanol were prepared. The biodiesel was prepared from RME and WAFME at a ratio of 1:1 (hereafter, this mixture is marked as R+W). The diesel and biodiesel blends contained 7, 10, 15 and 30 vol% R+W. Ranges of conditions were chosen for 7, 10, 30 vol%. The 7 vol% of biodiesel was chosen, for in EU this ratio is currently used diesel fuel B7, which can contain up to 7 vol% bio-component according to EN 590 to use. The quantity of 10 vol% was chosen as the next logical step for the future. This is the possible next target within the framework of ecology and sustainability of transport.

Table 1. Samples Composition

Name	Diesel (vol%)	RME (vol%)	WAFME (vol%)	Ethanol (vol%)	Butanol (vol%)
R+W7	93	3.5	3.5	0	0
R+W7BUT5	88	3.5	3.5	0	5
R+W7ET5	88	3.5	3.5	5	0
R+W7BUT10	83	3.5	3.5	0	10
R+W7ET10	83	3.5	3.5	10	0
R+W10	90	5	5	0	0
R+W10BUT5	85	5	5	0	5
R+W10ET5	85	5	5	5	0
R+W10BUT10	80	5	5	0	10
R+W10ET10	80	5	5	10	0
R+W15	85	7.5	7.5	0	0
R+W15BUT5	80	7.5	7.5	0	5
R+W15ET5	80	7.5	7.5	5	0
R+W15BUT10	75	7.5	7.5	0	10
R+W15ET10	75	7.5	7.5	10	0
R+W30	70	15	15	0	0
R+W30BUT5	65	15	15	0	5
R+W30ET5	65	15	15	5	0
R+W30BUT10	60	15	15	0	10
R+W30ET10	60	15	15	10	0
RME	0	100	0	0	0
WAFME	0	0	100	0	0
R+W	0	50	50	0	0
R+WBUT5	0	47.5	47.5	0	5
R+WET5	0	47.5	47.5	5	0
R+WBUT10	0	45	45	0	10
R+WET10	0	45	45	10	0
Ethanol	0	0	0	100	0
Butanol	0	0	0	0	100
Diesel	100	0	0	0	0

Concentrations of 15 and 30% are commonly tested and reported in publications dealing with similar topics (Pradelle *et al.* 2017; Devarajan *et al.* 2022; Reang *et al.* 2022). Furthermore, B15 and B30 is currently used in countries such as Indonesia or Brazil. The binary mixtures of diesel and biodiesel were also mixed with bio-alcohol in the amount of 5 or 10 vol%, resulting in ternary blends. The samples and their composition are shown in Table 1.

These low content bio-alcohols were chosen because maintaining a low cetane number of mixtures with a high content of alcohol is not feasible for diesel engines (Lapuerta *et al.* 2017). Diesel fuel is a complex mixture of different chemical compounds, from highly non-polar n-alkanes to moderately polar aromatic hydrocarbons. In alcohol-diesel blends, these aromatic structures may interact slightly with the hydroxyl group of the alcohol, forming transient dipoles (intermolecular forces weaker than the hydrogen bond). The attraction between the hydroxyl group of the alcohol molecule and the carboxyl group of the biodiesel fuel is stronger than that between the hydroxyl group and the aromatic hydrocarbons of diesel fuel (Lapuerta *et al.* 2015). Molecules of ethanol are polar by nature as it contains the hydroxyl (OH) group; however, ethanol has relatively, more robust lipophilicity compared to other higher alcohols due to its hydrophilic-lipophilic balance value (7.95), so, ethanol can be simply blended with diesel fuel (Liu *et al.* 2016). The solubility of ethanol and diesel depends upon the blend temperature and ethanol's purity (Liu *et al.*, 2016). Torres-Jimenez *et al.* (2011) reported a similar fact and concluded that at 30 °C temperature, the blend, which contains up to 15 vol% anhydrous ethanol (99.8 %), remains in a single phase for at least 4 weeks. Pradelle *et al.* (2017) noted that to increase the biodiesel concentration in commercial diesel fuel, higher concentrations of biodiesel are also a positive factor to stabilize the diesel–biodiesel–ethanol blends. Lee *et al.* (2017) was found that tri-fuel emulsion with 5% ethanol content was of the best in stability with little separation. Fernando and Hanna (2004) published the diesel–biodiesel–ethanol fuel blends are stable well below zero temperature and have equal or superior fuel properties to regular diesel fuel.

Methods

This research employed a fully factorial design for the experiment, where density and viscosity depend on the temperature and the concentration ratio of diesel–biodiesel–alcohol blends. The temperature limits were -10 and 60 °C and the highest possible ratios of biodiesel and/or alcohols in binary and ternary blends.

The selected physical properties and the methods of their measurement with regards to the fuels examined in the experiment are shown in Table 2. The values in this table were measured, the others were taken from Lapuerta *et al.* (2017).

Table 2. Used Fuels Properties and the Methods

Properties	Method	Diesel	RME	WAFME	Ethanol	Butanol
Purity (%, v/v)	-	-	-	-	99.8	99.8
Density at 15 °C (kg/m ³)	ISO 3675	0.834	0.875	0.872	0.791	0.809
Kinematic viscosity at 40 °C (mm ² /s)	ISO 3104	2.578	4.543	4.537	1.147	2.316
Pour point (°C)	-	-20	-11	11	-114.1	-89.8

Density and viscosity measuring methods

Two physical properties were measured, namely the density and dynamic viscosity. The density was measured with a Densito 30 PX densitometer by Mettler Toledo (Columbus, OH, USA). The density was measured at 0, 20, and 40 °C. From these values, the values of the density of the fuels and their mixtures in the used temperature range were calculated using linear regression. A DV2T rotary viscometer Brookfield (Toronto, Canada) was used to measure the dynamic viscosity. All the samples were measured in a temperature range from -10 to 60 °C, which corresponds to the climatic temperature range for most European countries.

The kinematic viscosity was determined by Eq. 1,

$$\nu = \frac{\eta}{\rho} \quad (1)$$

where η is the dynamic viscosity, and ρ is the volumetric mass density. Each viscosity value given in Table A3 is an arithmetic mean calculated from $N = 20$ measurements. The same procedure as in (Trost *et al.* 2021) was used in the measurement.

Equations for the density and viscosity modelling

A linear density model was created from the measured density values according to (Trost *et al.* 2021) for further use, which is used to predict the density depending on the temperature in the range chosen:

$$\rho = \rho_0 \cdot T + q \quad (2)$$

In this equation, ρ is the mass density ($\text{g}\cdot\text{cm}^{-3}$), ρ_0 is the reference value of the mass density ($\text{g}\cdot\text{cm}^{-3}\cdot\text{K}^{-1}$), T is the absolute temperature (K), and q is the coefficient ($\text{g}\cdot\text{cm}^{-3}$).

Four models that are used in the literature were chosen to predict the viscosity-temperature behaviour of the fuels. The power law model was used by Kanaveli *et al.* (2017) for the prediction of diesel-AFME mixtures (made from corn, Jatropha, pomace, sesame, soy, sunflower and waste frying oil). The exponential model was used to predict ternary blends (palm biodiesel-butanol-waste plastic oil) in the article by Mujtaba *et al.* (2021). Lapuerta *et al.* (2017) used the Arrhenius model for the prediction of ternary blends of diesel-FAME (soybean and palm oil)-butanol and ethanol blends. In another paper (Kanaveli *et al.* 2017), the Arrhenius model was used to predict diesel-FAME blends. Vogel's model was used in publications (Gülüm and Bilgin 2018; Gülüm *et al.* 2018) for diesel-WAFME-alcohol (methanol, ethanol, isopropanol, butanol and pentanol) ternary blends.

Four appropriate mathematical models were chosen to predict the viscosity-temperature behaviour of the fuels and their mixtures,

$$\text{Power law } \nu = \nu_{0p} \cdot T^a \quad (3)$$

$$\text{Exponential } \nu = \nu_0 \cdot e^{b \cdot T} \quad (4)$$

$$\text{Arrhenius } \nu = \nu_0 \cdot e^{\frac{E_a}{R \cdot T}} \quad (5)$$

$$\text{Vogel } \nu = \nu_0 \cdot e^{\frac{c}{d+T}} \quad (6)$$

where ν is the kinematic viscosity ($\text{mm}^2\cdot\text{s}^{-1}$), ν_{0p} is the reference value of the Power law kinematic viscosity ($\text{mm}^2\cdot\text{s}^{-1}\cdot\text{K}^{-a}$), a is the Power law coefficient (–), ν_0 is the reference value of the kinematic viscosity ($\text{mm}^2\cdot\text{s}^{-1}$), b is the exponential coefficient (K^{-1}), c and d

are the Vogel coefficients (K), T is the absolute temperature (K), E_a is the Arrhenius activation energy ($\text{J}\cdot\text{mol}^{-1}$), and R is the Universal gas constant ($\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$).

Data processing

The software packages Matlab® (Natick, MA, USA) and Statistica® (Palo Alto, CA, USA) were used to process and evaluate the measured data. To determine the statistically significant differences in the viscosity of the fuel mixtures, an analysis of variance (ANOVA) test was used with multiple comparisons using Tukey's HSD (Honestly Significant Difference) test at a significance level of $p < 0.05$.

The appropriateness of using the model describing the dependence of the kinematic viscosity on the temperature of ternary blends was expressed by the basic regression characteristics - coefficient of determination (R^2) and the sum of the squared estimate of errors (SSE).

RESULTS AND DISCUSSION

Dependence of the Density on the Temperature

The density values of the ternary blends are given in Table A1 (see Appendix). It is evident from the measured results that the temperature had a significant effect on the density of the ternary blends and individual fuels. With an increasing temperature, a linear decrease in the density of all the ternary blends and individual fuels can be observed. The same conclusions were also observed by the authors Chhetri and Watts (2012) when monitoring the density of biodiesel produced from rapeseed. Other authors observed a linear behaviour of the density depending on the temperature of the biodiesel-diesel-bio-alcohol (Isopropyl alcohol and 1-butyl alcohol) mixtures (Nita and Geacai 2013). The greater the amount of biodiesel contained in the mixture, the higher the density of the mixture (see Table A1). This result is confirmed by Prietoa *et al.* (2015), where they state that the density of cottonseed biodiesel has a linearly negative dependence on the temperature and a linearly positive dependence on the increasing content of the unsaturated fats. In contrast, with a higher amount of alcohol in the mixture, the density decreases. Similar conclusions were reached in a paper (Razzaq *et al.* 2020), where the authors report that the density of binary biodiesel blends decreases with the addition of bioethanol. Furthermore, it can be observed that the greater the amount of biodiesel in the mixture, the greater the effect of the alcohol on the density reduction.

The quality of the proposed linear model was assessed using two basic statistical characteristics - the coefficient of determination (R^2) and the Error Sum of the Squares (SSE), where R^2 is equal to 0.9993 and SSE is equal to $9.52\cdot 10^{-7}$. The determination coefficient R^2 was also used to determine the accuracy of the mathematical model of the biodiesel density behaviour in the article by Yuan *et al.* (2009). In the same paper, it was found that the density of biodiesel as a function of temperature is linear. A complete overview of the coefficients of the linear model describing the dependence of the density on the temperature of ternary blends is presented in Table A2.

Dependence of the Viscosity on the Temperature

The temperature had a significant effect on the viscosity of the ternary blends, which can be seen in Fig. 1 and using the values given in Table A3. The kinematic viscosity of a mixture of two methyl esters and diesel decreases with an increasing temperature,

which was published by Hosamani *et al.* (2022). Figure 1 shows that the viscosity of the ternary blends increased linearly with the increasing proportion of R+W in the mixture. A multi-dimensional polynomial model was created from the measured viscosity values of the R+W fuel mixture,

$$\nu(T, C) = 1138 - 10.77 \cdot T + 1.912 \cdot C + 0.03417 \cdot T^2 - 0.01162 \cdot T \cdot C - 3.625 \cdot 10^{-5} \cdot T^3 + 1.781 \cdot 10^{-5} \cdot T^2 \cdot C \quad (7)$$

where ν is the kinematic viscosity ($\text{mm}^2 \cdot \text{s}^{-1}$), T is the absolute temperature (K), and C is the concentration (vol%). All the coefficients were statistically significant ($R^2 = 0.9978$, $\text{SSE} = 1.895$).

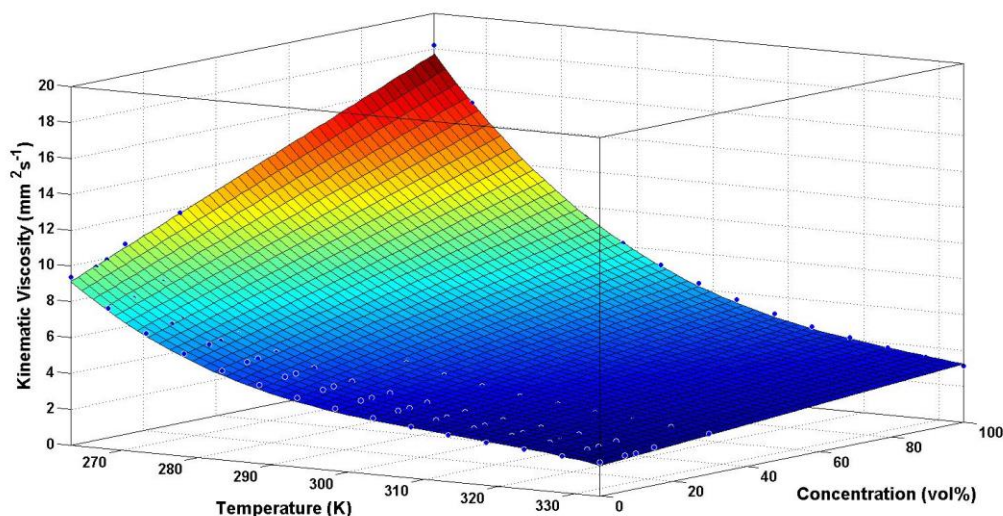


Fig. 1. Three-dimensional graph created using a multivariate polynomial model showing the effect of the temperature and the R+W concentration in diesel

The increasing amount of bio-alcohol in the ternary blends caused a decrease in the kinematic viscosity (see Table A3). Similar conclusions were also reached by Lapuerta *et al.* (2017) and Razzaq *et al.* (2020), who found that increasing alcohol content contributed to a non-linear decrease in the viscosity of the three-component mixture of biofuels in the composition of diesel-biodiesel-ethanol. Lee *et al.* (2017) reported that an emulsion of 85% diesel, 10% biodiesel, and 5% ethanol appears to be the optimal alternative fuel. The higher the R+W ratio in the mixture, the more the bio-alcohol reduces the viscosity of the mixture. A more pronounced reduction in the viscosity was observed after the addition of bio-alcohol with a shorter carbon chain – bioethanol. A similar behaviour of the kinematic viscosity of ternary blends was observed by Nita and Geacai (2013) and for two-component mixtures (Chhetri and Watts 2012; Lapuerta *et al.* 2017). The investigation of the kinematic viscosity of WAFME from various sources was dealt with by Sander *et al.* (2018). The authors reported that the viscosity of WAFME varies depending on the feedstock. The lowest biodiesel viscosity was observed with synthesized chicken fat (Sander *et al.* 2018). The influence of the kinematic viscosity of ternary blends by higher concentrations of biodiesel and bioethanol was shown by Saleh and Selim (2017). The authors (Al-Esawi *et al.* 2019) reported that a blend of 15% biodiesel, 5% ethanol, and 80% diesel showed a viscosity reduction of less than 2% compared to that of pure diesel.

Using the ANOVA test, through multiple comparisons, it was found that if one were to add the same amount of bioethanol to the mixture with the lowest R+W content (7 vol%) instead of 5 vol% biobutanol, there would be a statistically significant change in the kinematic viscosity (at a significance level of 95 %), or the kinematic viscosity of low R+W fuel is influenced by the type of bio-alcohol added.

If the R+W content in the ternary blends increases above 10 vol%, then the influence of the type of bio-alcohol on the change in the kinematic viscosity decreases. The results of the ANOVA tests revealed that the kinematic viscosity changed statistically significantly according to the type of added bio-alcohol (at a concentration of 5 vol%) only at lower fuel temperatures, as discussed below. A statistically significant effect of the type of bio-alcohol on the change in the kinematic viscosity was observed for the R+W10 mixture from -10 to 30 °C and for the R+W15 mixture only from -10 to 20 °C. For the ternary blends, in which the proportion of R+W was 30 vol%, the type of added bio-alcohol (in the amount of 5 vol%) did not statistically significantly affect the kinematic viscosity at temperatures of 10 °C, 5 °C, 0 °C and -5 °C. The statistical evaluation also showed that the type of bio-alcohol significantly affects the viscosity of the R+W mixture if its amount is increased to 10 vol% in the ternary blends.

It also follows from the measured data that with the decreasing temperature of the mixtures, larger differences in the kinematic viscosity are evident between the individual types of evaluated fuel mixtures. The growing difference between the kinematic viscosity values of the individual mixtures with a decreasing temperature was also evident from the expressed temperature parameters of the model calculated using ANOVA (Table 3). As shown, the estimated parameters of the ANOVA tests decreased with an increasing temperature (thus, also the effect of the factor “Temperature”). With higher temperatures, the differences between the kinematic viscosity of the individual fuels will be lower.

Table 3. Parameter Estimates of the ANOVA (Over-parameterized model)

Effect	Level	Comment	Kin. Viscosity Parameter	Kin. Viscosity Std. Err.
Intercept			1.540667	0.051066
Temperature (°C)	-10	Biased	6.872021	0.072219
	-5	Biased	5.294708	0.072219
	0	Biased	4.208227	0.072256
	5	Biased	3.372791	0.072256
	10	Biased	2.708125	0.072219
	15	Biased	2.176000	0.072219
	20	Biased	1.752444	0.072256
	25	Biased	1.397872	0.072256
	30	Biased	1.091354	0.072219
	35	Biased	0.835417	0.072219
	40	Biased	0.610417	0.072219
	45	Biased	0.414917	0.072219
	50	Biased	0.241812	0.072219
	55	Biased	0.103979	0.072219
60	Zeroed*		0.000000	

*Zeroed predictors failed tolerance check

Based on these findings, the rate of the decrease in the kinematic viscosity of the given fuel will decrease as the temperature increases. By creating a R+W mixture in a 1:1 ratio, the pour point was significantly reduced by 7.5 °C (from 11 to 3.5 °C) compared to pure WAFME.

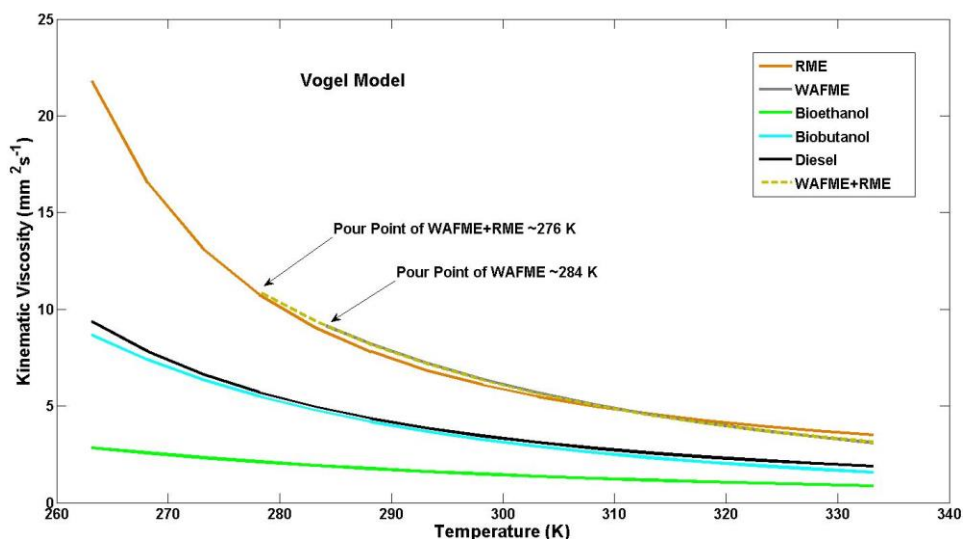


Fig. 2. Comparison of the temperature behavior and pour point of pure fuels

The original viscosity value for the R+W mixture in a 1:1 ratio at 40 °C was 4.561 mm²/s. The addition of bio-alcohol resulted in an improvement in the flow properties and a reduction in the pour point of R+W (see Table A6), and illustratively for R+W30 mixtures, see Fig. 3. By adding 5 vol% biobutanol, the viscosity decreased to a value of 4.131 mm²/s, whereas by adding 5 vol% of bioethanol, the viscosity then decreased to 4.059 mm²/s. Similarly, with the addition of 10 vol% biobutanol, the viscosity dropped to a value of 3.865 mm²/s, and with the addition of 10 vol% bioethanol, the viscosity then decreased to 3.774 mm²/s. The pour point was 2.3 °C with the addition of 5 vol% butanol, whereas the pour point was 2.2 °C with the addition of 5 vol% bioethanol. In the case of 10 vol% biobutanol and bioethanol, the pour point was identically 1.4 °C.

The pour point of a mixture of diesel fuel and biodiesel is influenced by the amount of fatty acids contained in the mixture (Nita and Geacai 2013). Golimowski *et al.* (2017) found that the cold filter plugging point (CFPP) value, which is closely related to the pour point, is directly proportional to the increase in saturated fatty acid content. The diesel dilution improves the low-temperature flow properties of a biodiesel (Bhale *et al.* 2009; Ramalho *et al.* 2012). However, this process is only applicable up to 30 vol% biodiesel (Nita and Geacai 2013). A pour point RME reduction was also achieved by adding cold flow improvers (CFIs) in the form of sorbitan derivatives and palmitate derivatives, with Span 40 being the most effective (Abe *et al.* 2021). According to Imtenan *et al.* (2015), the pour point of WAFME was observed at 8 °C, which is also close to our findings. The difference could be due to the different raw material for the production of WAFME (Golimowski *et al.* 2017). This is also stated by Monirul *et al.* (2015) that the properties of biodiesel and oxidation stability are dependent on the composition of fatty acids.

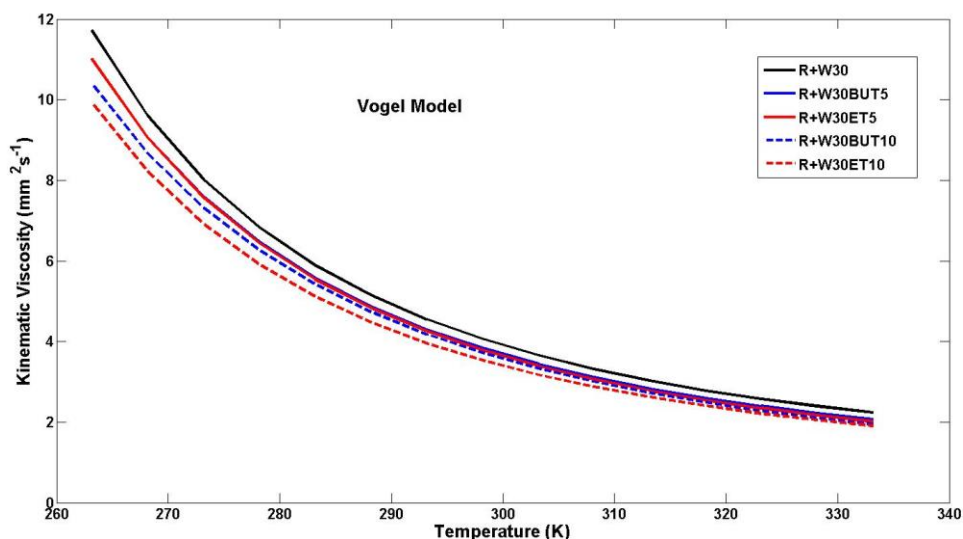


Fig. 3. Comparison of the alcohol impact on the R+W30 blend viscosity by using the Vogel model

Predictive Mathematical Models

The dependence of the kinematic viscosity of the ternary blends on the temperature was modelled using four mathematical models. A comparison of the models for the mixture R+W10ET5 and R+W10BUT5 can be seen in Fig. 4. Complete tables of all the coefficients of the used mathematical models for the viscosity-temperature behaviour of ternary blends are presented in Tables A4 through A7.

Table 4 shows the average values of the basic statistical characteristics, through which, as already mentioned, the assessment of the quality of the proposed models (power law, exponential, Arrhenius, Vogel) for the viscosity-temperature behaviour of the ternary blends was conducted. The R^2 determination coefficients for all the selected models achieved high values.

Table 4. Evaluation of the Viscosity Predictive Models

Predictive Viscosity Models	SSE	R^2
Power law	1.0624	0.9909
Exponential	1.5901	0.9850
Arrhenius	0.6456	0.9951
Vogel	0.0621	0.9997

Vogel's model was used to predict the binary and ternary blends (waste cooking oil-diesel-alcohol) by Gülüm and Bilgin (2018). As in the present work, Vogel's model was found to explain a high percentage of the information contained in the data. The same authors also point out that the Arrhenius model achieves higher errors than Vogel's model (Gülüm and Bilgin 2018). Gülüm *et al.* (2018) confirm that Vogel's model qualitatively accurately corresponds to the data on the viscosity of the RME and diesel fuel mixtures. According to Gülüm and Bilgin (2018), in addition to the Vogel model, the Rational model also achieved a high prediction value for ternary blends (waste cooking oil-diesel-alcohol).

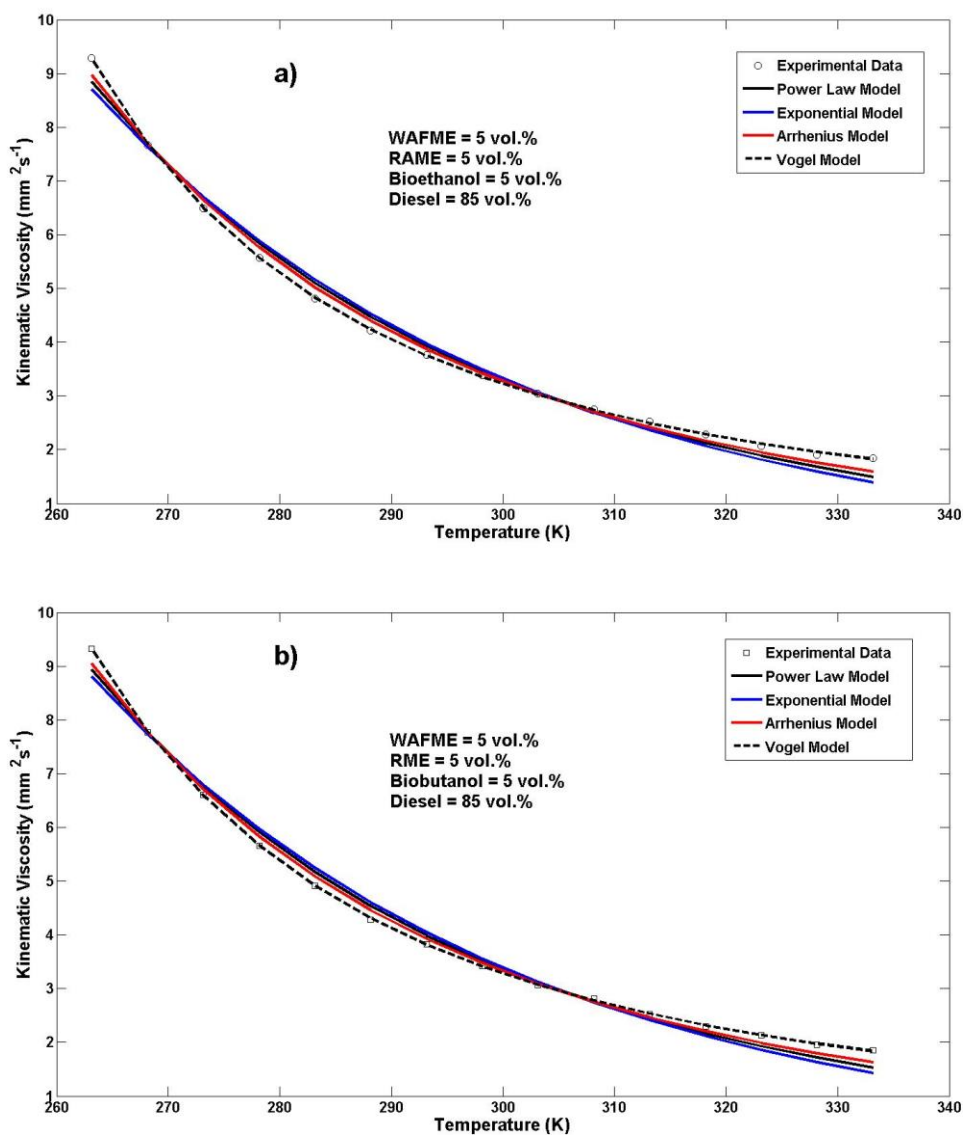


Fig. 4. Comparison of the predictive mathematical models used for (a) R+W10ET5 and (b) R+W10BUT5

CONCLUSIONS

1. The density of ternary blends in a temperature range from -10 to 60 °C decreased linearly with an increasing temperature.
2. The kinematic viscosity of the ternary blends in a temperature range from -10 to 60 °C decreased non-linearly with an increasing temperature. The kinematic viscosity of the ternary blends increased with increasing proportion of waste animal fatty methyl ester (WAFME) at a ratio of 1:1 (R+W). The addition of bio-alcohol (5 or 10 vol%) had an effect on reducing the overall kinematic viscosity of the ternary blends.

3. Mixing R+W in a ratio of 1:1 reduced the pour point by 7.5 °C (from 11 to 3.5 °C) compared to pure WAFME. The addition of bioethanol resulted in a higher decrease in the viscosity of the ternary blends than in the case of the addition of biobutanol. In the case of the addition of 5 vol% bioethanol, the R+W pour point decreased to 2.2 °C, whereas in the case of the addition of 10 vol% bioethanol the pour point then decreased to 1.4 °C.
4. Of the four mathematical models used for predicting the viscosity-temperature behaviour, Vogel's model was the most accurate (SSE = 0.0621, R² = 0.9997).
5. The research hypothesis was disproved. Through exact measurements and the subsequent modelling, it was determined that using a suitable ratio of a mixture of diesel fuel, WAFME, rapeseed methyl ester (RME), and bioethanol or biobutanol, it was not possible to achieve the same flow properties (including the pour point) as pure diesel fuel, or diesel with the mandatory addition of biodiesel, but they can be approached quite well, which could be found to be sufficient in some cases of use.

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APPENDIX

Table A1. Density of the Ternary Blends in g/cm³ (Mean Density Values of the Fuels are given, N = 5)

Temperature [°C]	-10	-5	0	5	10	15	20	25	30	35	40	45	50	55	60
Temperature [K]	263.15	268.15	273.15	278.15	283.15	288.15	293.15	298.15	303.15	308.15	313.15	318.15	323.15	328.15	333.15
R+W7	0.846	0.844	0.841	0.839	0.837	0.834	0.832	0.830	0.827	0.825	0.823	0.82	0.818	0.816	0.813
R+W7BUT5	0.845	0.842	0.84	0.837	0.835	0.832	0.83	0.827	0.825	0.822	0.82	0.817	0.815	0.812	0.81
R+W7ET5	0.845	0.842	0.84	0.837	0.835	0.832	0.83	0.827	0.825	0.822	0.82	0.817	0.815	0.812	0.81
R+W7BUT10	0.845	0.842	0.839	0.837	0.834	0.831	0.829	0.826	0.824	0.821	0.818	0.816	0.813	0.81	0.808
R+W7ET10	0.843	0.840	0.838	0.835	0.832	0.830	0.827	0.825	0.822	0.819	0.817	0.814	0.811	0.809	0.806
R+W10	0.847	0.845	0.842	0.840	0.838	0.835	0.833	0.830	0.828	0.826	0.823	0.821	0.819	0.816	0.814
R+W10BUT5	0.846	0.843	0.841	0.838	0.836	0.833	0.831	0.828	0.826	0.823	0.821	0.818	0.816	0.813	0.811
R+W10ET5	0.843	0.841	0.839	0.837	0.834	0.832	0.830	0.828	0.825	0.823	0.821	0.819	0.816	0.814	0.812
R+W10BUT10	0.844	0.841	0.839	0.837	0.834	0.832	0.829	0.827	0.825	0.822	0.820	0.818	0.815	0.813	0.810
R+W10ET10	0.843	0.841	0.838	0.836	0.834	0.831	0.829	0.826	0.824	0.822	0.819	0.817	0.815	0.812	0.810
R+W15	0.849	0.847	0.845	0.842	0.840	0.838	0.835	0.833	0.830	0.828	0.826	0.823	0.821	0.819	0.816
R+W15BUT5	0.846	0.844	0.842	0.840	0.837	0.835	0.833	0.831	0.828	0.826	0.824	0.822	0.819	0.817	0.815
R+W15ET5	0.846	0.844	0.842	0.839	0.837	0.835	0.833	0.831	0.829	0.827	0.825	0.822	0.820	0.818	0.816
R+W15BUT10	0.845	0.843	0.841	0.839	0.837	0.835	0.833	0.831	0.829	0.827	0.825	0.823	0.821	0.819	0.817
R+W15ET10	0.843	0.841	0.839	0.837	0.835	0.833	0.831	0.830	0.828	0.826	0.824	0.822	0.820	0.818	0.816
R+W30	0.855	0.853	0.851	0.849	0.846	0.844	0.842	0.840	0.837	0.835	0.833	0.831	0.828	0.826	0.824
R+W30BUT5	0.855	0.852	0.850	0.847	0.844	0.842	0.839	0.837	0.834	0.831	0.829	0.826	0.823	0.821	0.818
R+W30ET5	0.854	0.851	0.848	0.846	0.843	0.840	0.838	0.835	0.833	0.830	0.827	0.825	0.822	0.819	0.817
R+W30BUT10	0.854	0.851	0.849	0.846	0.844	0.841	0.839	0.836	0.834	0.831	0.829	0.826	0.824	0.821	0.819
R+W30ET10	0.851	0.849	0.846	0.844	0.842	0.839	0.837	0.834	0.832	0.830	0.827	0.825	0.823	0.820	0.818
RME	0.887	0.884	0.882	0.880	0.878	0.875	0.873	0.871	0.869	0.866	0.864	0.862	0.860	0.857	0.855
WAFME					PP +11	0.872	0.870	0.867	0.865	0.862	0.860	0.857	0.855	0.852	0.850
R+W			PP +3.5	0.879	0.876	0.874	0.871	0.868	0.866	0.863	0.860	0.858	0.855	0.852	0.850
R+WBUT5			PP +2.3	0.878	0.875	0.872	0.868	0.865	0.862	0.859	0.856	0.853	0.850	0.847	0.844
R+WET5			PP +2.2	0.876	0.874	0.871	0.868	0.865	0.862	0.859	0.856	0.853	0.850	0.847	0.844
R+WBUT10			PP +1.4	0.871	0.869	0.867	0.864	0.862	0.859	0.857	0.855	0.852	0.850	0.847	0.845
R+WET10			PP +1.4	0.874	0.871	0.868	0.865	0.862	0.859	0.856	0.853	0.850	0.847	0.844	0.841
Ethanol	0.805	0.802	0.799	0.797	0.794	0.791	0.789	0.786	0.784	0.781	0.778	0.776	0.773	0.770	0.768
Butanol	0.819	0.817	0.815	0.813	0.811	0.809	0.807	0.805	0.803	0.801	0.799	0.797	0.795	0.793	0.791
Diesel	0.846	0.843	0.841	0.839	0.837	0.834	0.832	0.830	0.828	0.825	0.823	0.821	0.819	0.816	0.814

Table A2. Coefficients and Statistical Indicators (R^2 – Coefficient of Determination, SSE – Sum of the squared estimate of errors) of the Linear Model

Linear	q (g·cm ⁻³)	ρ_o (g·cm ⁻³ ·K ⁻¹)	SSE	R^2
R+W7	-0.0004679	0.9692	1.101e-06	0.9993
R+W7BUT5	-0.0005	0.9763	9.333e-07	0.9995
R+W7ET5	-0.0005	0.9763	9.333e-07	0.9995
R+W7BUT10	-0.0005271	0.9834	1.243e-06	0.9994
R+W7ET10	-0.0005257	0.9813	1.105e-06	0.9994
R+W10	-0.0004743	0.9719	1.105e-06	0.9993
R+W10BUT5	-0.0005	0.9773	9.333e-07	0.9995
R+W10ET5	-0.0004486	0.9613	1.086e-06	0.9992
R+W10BUT10	-0.0004771	0.9693	1.276e-06	0.9992
R+W10ET10	-0.0004743	0.9679	1.105e-06	0.9993
R+W15	-0.0004729	0.9738	1.243e-06	0.9992
R+W15BUT5	-0.0004486	0.9643	1.086e-06	0.9992
R+W15ET5	-0.0004286	0.9587	1.219e-06	0.9991
R+W15BUT10	-0.0004	0.9503	3.698e-31	0.9999
R+W15ET10	-0.00038	0.9428	9.333e-07	0.9991
R+W30	-0.0004486	0.9733	1.086e-06	0.9992
R+W30BUT5	-0.0005257	0.9933	1.105e-06	0.9994
R+W30ET5	-0.0005271	0.9924	1.243e-06	0.9994
R+W30BUT10	-0.0005	0.9853	9.333e-07	0.9995
R+W30ET10	-0.0004743	0.9759	1.105e-06	0.9993
RME	-0.0004514	1.005	1.219e-06	0.9991
WAFME	-0.000497	1.015	6.061e-07	0.9988
R+W	-0.0005315	1.027	8.765e-07	0.9991
R+WBUT5	-0.0006189	1.05	9.755e-07	0.9993
R+WET5	-0.0005923	1.041	7.051e-07	0.9994
R+WBUT10	-0.0004797	1.005	9.464e-07	0.9989
R+WET10	-0.0006	1.041	7.149e-31	0.9999
Ethanol	-0.0005271	0.9434	1.243e-06	0.9994
Butanol	-0.0004	0.9243	2.465e-32	0.9999
Diesel	-0.0004514	0.9645	1.219e-06	0.9991
Average value: SSE = 9.52E-07, R^2 = 0.9993				

Temperature [°C]	R+W7	R+W7BUT5	R+W7ET5	R+W7BUT10	R+W7ET10	R+W10	R+W10BUT5	R+W10ET5	R+W10BUT10	R+W10ET10	R+W15	R+W15BUT5	R+W15ET5	R+W15BUT10	R+W15ET10	R+W30	R+W30BUT5	R+W30ET5	R+W30BUT10	R+W30ET10	RME	WAFME	R+W	R+WBUT5	R+WET5	R+WBUT10	R+WET10	Ethanol	Butanol	Diesel
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Table A4. Values of the Coefficients of and Statistical Indicators for the Power Law Model (R^2 – Coefficient of Determination, SSE – Sum of Squared Estimate of Errors)

Power Law	v_{op} (mm ² ·s ⁻¹ ·K ^{-a})	a (-)	SSE	R ²
R+W7	5.621e+18*	-7.348	0.6481	0.9916
R+W7BUT5	1.244e+19*	-7.499	0.8239	0.9884
R+W7ET5	1.112e+19*	-7.483	0.6749	0.9901
R+W7BUT10	7.768e+18*	-7.422	0.5429	0.9916
R+W7ET10	7.918e+18*	-7.434	0.6847	0.9885
R+W10	1.181e+19*	-7.476	0.7436	0.991
R+W10BUT5	1.102e+19*	-7.475	0.6327	0.9913
R+W10ET5	1.466e+19*	-7.528	0.7321	0.9897
R+W10BUT10	1.661e+19*	-7.554	0.6826	0.9902
R+W10ET10	8.067e+18*	-7.437	0.8034	0.9863
R+W15	3.415e+19*	-7.657	1.3820	0.9851
R+W15BUT5	1.609e+19*	-7.535	0.8453	0.9894
R+W15ET5	2.030e+19*	-7.578	0.7733	0.9902
R+W15BUT10	1.611e+19*	-7.543	0.7402	0.9899
R+W15ET10	1.272e+19*	-7.510	0.7611	0.9884
R+W30	7.458e+19*	-7.779	1.5160	0.9867
R+W30BUT5	8.257e+19*	-7.808	1.1680	0.9886
R+W30ET5	1.602e+20*	-7.927	1.079	0.9896
R+W30BUT10	4.600e+19*	-7.711	0.7123	0.9924
R+W30ET10	1.602e+20*	-7.927	1.0790	0.9896
RME	4.532e+23*	-9.238	13.11	0.9674
WAFME	5.002e+17*	-6.826	0.03847	0.9985
R+W	4.21e+18*	-7.198	0.2379	0.9966
R+WBUT5	2.00e+18*	-7.084	0.2042	0.9963
R+WET5	3.618e+18*	-7.192	0.1738	0.9968
R+WBUT10	1.855e+18*	-7.083	0.1260	0.9974
R+WET10	1.850e+18*	-7.089	0.0973	0.9978
Ethanol	5.718e+12	-5.085	0.0066	0.9988
Butanol	1.658e+19*	-7.557	0.1687	0.9975
Diesel	7.175e+18*	-7.397	0.6847	0.9906

*Coefficient is not statistically significant at a confidence level of $p < 0.05$. Average value: SSE = 1.0624, $R^2 = 0.9909$

Table A5. Values of the Coefficients and Statistical Indicators for the Exponential Model (R^2 – Coefficient of Determination, SSE – Sum of the Squared Estimate of Errors)

Exponential	v_0 (mm ² ·s ⁻¹)	b (K ⁻¹)	SSE	R^2
R+W7	3.746	-0.56970	1.128	0.9855
R+W7BUT5	8169	-0.02601	1.322	0.9814
R+W7ET5	7028	-0.02577	1.102	0.9814
R+W7BUT10	7286	-0.02574	0.941	0.9855
R+W7ET10	7028	-0.02577	1.102	0.9814
R+W10	8629	-0.02593	1.265	0.9847
R+W10BUT5	8090	-0.02593	1.088	0.9850
R+W10ET5	8406	-0.02612	1.208	0.9831
R+W10BUT10	8473	-0.02621	1.133	0.9837
R+W10ET10	7018	-0.02578	1.244	0.9789
R+W15	10732	-0.02657	2.098	0.9773
R+W15BUT5	3.679	-0.5845	1.382	0.9826
R+W15ET5	9259	-0.0263	1.280	0.9839
R+W15BUT10	8655	-0.02617	1.229	0.9833
R+W15ET10	7901	-0.02605	1.208	0.9815
R+W30	13350	-0.02701	2.358	0.9794
R+W30BUT5	12972	-0.02712	1.871	0.9817
R+W30ET5	14570	-0.02756	1.768	0.9829
R+W30BUT10	11361	-0.02679	1.255	0.9866
R+W30ET10	10112	-0.02656	1.404	0.983
RME	9.519e+04*	-0.03226	16.97	0.9577
WAFME	4901	-0.02224	0.0845	0.9968
R+W	8432	-0.02401	0.4598	0.9927
R+WBUT5	6829	-0.02362	0.3941	0.9929
R+WET5	7447	-0.02399	0.3467	0.9936
R+WBUT10	6385	-0.02362	0.2685	0.9945
R+WET10	6189	-0.02364	0.2178	0.9952
Ethanol	277.2	-0.01748	0.01882	0.9965
Butanol	8403	-0.02626	0.3964	0.9941
Diesel	7548	-0.02565	1.159	0.9841

*Coefficient is not statistically significant at a confidence level of $p < 0.05$. Average value: SSE = 1.5901, $R^2 = 0.9850$

Table A6. Values of the Coefficients and Statistical Indicators for the Arrhenius Model (R^2 – Coefficient of Determination, SSE – Sum of the Squared Estimate of Errors)

Arrhenius	v_0 (mm ² ·s ⁻¹)	E_a (J·mol ⁻¹)	SSE	R^2
R+W7	0.003079	1.756e+04	0.3020	0.9961
R+W7BUT5	0.002494	1.791e+04	0.4453	0.9937
R+W7ET5	0.002478	1.787e+04	0.3472	0.9949
R+W7BUT10	0.002592	1.773e+04	0.2556	0.9961
R+W7ET10	0.002445	1.776e+04	0.3676	0.9938
R+W10	0.002756	1.785e+04	0.3626	0.9956
R+W10BUT5	0.002589	1.785e+04	0.3009	0.9959
R+W10ET5	0.002419	1.798e+04	0.3768	0.9947
R+W10BUT10	0.002318	1.804e+04	0.3483	0.9950
R+W10ET10	0.002422	1.778e+04	0.4616	0.9922
R+W15	0.002385	1.829e+04	0.8196	0.9911
R+W15BUT5	0.002532	1.800e+04	0.4419	0.9944
R+W15ET5	0.002413	1.809e+04	0.3989	0.9950
R+W15BUT10	0.002420	1.801e+04	0.3777	0.9949
R+W15ET10	0.002356	1.794e+04	0.4225	0.9935
R+W30	0.002323	1.856e+04	0.8633	0.9925
R+W30BUT5	0.002132	1.863e+04	0.6336	0.9938
R+W30ET5	0.001887	1.89e+04	0.5616	0.9946
R+W30BUT10	0.002266	1.839e+04	0.3256	0.9965
R+W30ET10	0.002274	1.826e+04	0.4452	0.9946
RME	0.0008934*	2.194e+04	9.8050	0.9756
WAFME	0.005765	1.738e+04	0.01301	0.9995
R+W	0.004708	1.79e+04	0.09338	0.9987
R+WBUT5	0.004791	1.762e+04	0.07689	0.9986
R+WET5	0.004207	1.788e+04	0.06179	0.9989
R+WBUT10	0.004491	1.761e+04	0.0382	0.9992
R+WET10	0.004299	1.763e+04	0.02723	0.9994
Ethanol	0.010600	1.224e+04	0.004682	0.9991
Butanol	0.002279	1.802e+04	0.05491	0.9992
Diesel	0.002822	1.767e+04	0.3357	0.9954

*Coefficient is not statistically significant at a confidence level of $p < 0.05$. Average value: SSE = 0.6456, $R^2 = 0.9951$

Table A7. Values of the Coefficients and Statistical Indicators for the Vogel Model (R^2 – Coefficient of Determination, SSE – Sum of the Squared Estimate of Errors)

Vogel	v_0 (mm ² ·s ⁻¹)	c (K)	d (K)	SSE	R ²
R+W7	0.1412	485.0	- 148.4	0.006725	0.9999
R+W7BUT5	0.1970	375.3	- 165.8	0.01360	0.9998
R+W7ET5	0.1580	425.1	- 158.1	0.01814	0.9997
R+W7BUT10	0.1263	483.8	- 149.2	0.006742	0.9999
R+W7ET10	0.1843	374.2	- 165.5	0.007622	0.9999
R+W10	0.1517	462.0	- 152.7	0.012750	0.9998
R+W10BUT5	0.1368	472.6	- 151.2	0.004225	0.9999
R+W10ET5	0.1651	416.7	- 159.7	0.01050	0.9999
R+W10BUT10	0.1531	429.9	- 157.9	0.01333	0.9998
R+W10ET10	0.2240	327.6	- 173.1	0.02146	0.9996
R+W15	0.2876	308.1	- 177.8	0.03335	0.9996
R+W15BUT5	0.1830	403.8	- 161.7	0.01204	0.9998
R+W15ET5	0.1555	441.0	- 156.5	0.03224	0.9996
R+W15BUT10	0.1549	435.8	- 157.0	0.02055	0.9997
R+W15ET10	0.2240	327.6	- 173.1	0.02146	0.9996
R+W30	0.2621	344.1	- 172.6	0.02799	0.9998
R+W30BUT5	0.2025	389.1	- 165.8	0.02257	0.9998
R+W30ET5	0.1696	425.0	- 161.3	0.01260	0.9999
R+W30BUT10	0.1135	536.1	- 144.5	0.01557	0.9998
R+W30ET10	0.1706	416.8	- 160.6	0.01492	0.9998
RME	0.8601	173.4	-209.5	1.44	0.9964
WAFME	0.03666*	1111	-82.83	0.007449	0.9997
R+W	0.0931	737.4	- 123.4	0.019390	0.9997
R+WBUT5	0.1095	660.4	- 131.5	0.002073	1
R+WET5	0.08014	749.1	- 122	0.005833	0.9999
R+WBUT10	0.05861	860.1	- 108.1	0.004204	0.9999
R+WET10	0.04017	993.8	- 93.96	0.006727	0.9999
Ethanol	0.003907*	2108	56.76*	0.004131	0.9992
Butanol	0.01055*	1377	-57.97	0.0407	0.9994
Diesel	0.1565	444.2	- 154.6	0.005392	0.9999

*Coefficient is not statistically significant at a confidence level of $p < 0.05$. Average value: SSE = 0.0621, $R^2 = 0.9997$