

Predicting the Temperature and Composition – Dependent Density and Viscosity of Diesel Fuel – Ethanol Blends

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Abstract

Density and viscosity are very important fuel properties which have a major influence not only on the fuel production, transportation and distribution processes but also on the processes that take place in an internal combustion engine. Developing robust and high precision density and viscosity models for stabilized diesel fuel – ethanol blends helps the production of fuel to adhere to the quality requirements regarding density and viscosity and the modeling and simulation of injection and combustion processes. For modeling the density and the viscosity of diesel fuel – ethanol blends, five mixtures were prepared with ethanol content up to 15 % (v/v) and were stabilized by adding tetrahydrofuran as a surfactant at room temperature. The temperature-dependent density and viscosity of the blends were measured at four different temperatures (0, 15, 40 and 50 °C) using an SVM 3000 type apparatus. Based on experimental data, several mixing rules were fitted to them and three new models were developed, of which two need only one experimental value. These models yield very good accuracies, presenting average relative deviations of 0.0604 % in the case of density and 3.8931 % in the case of viscosity.

Keywords

diesel – ethanol fuel blends, stable ternary blends, mathematical models, density, viscosity

1 Introduction

Nowadays, transportation is one of the most pollutant sector in urban environment because of the fossil nature of the energy source and the process of transformation of the chemical energy into mechanical work [1]. Another concern is the petrol reserves which are more and more limited and expensive [2]. On the one hand, in spite of the current development of alternative drive-trains of the automotive vehicles (hybrid, plug-in hybrid and electric), internal fossil-fueled combustion engines (ICE) will remain the most used propellant [3] for a while, even if they are banned in large urban agglomerations, like Stuttgart, Hamburg, Paris, Madrid, Paris, Madrid, Copenhagen, etc. [4]. On the other hand, even if there are several technological approaches to reduce the fuel consumption and environmental pollution caused by fossil fuels, they have their costs on individuals and the society as well, which is a rational contradiction between transportation and environmental protection [5]. Meanwhile, identifying more environmentally friendly fuels for ICE is the key for the transition from fossil fuels to electricity or

some combination of them. Blends of diesel fuel – ethanol appear to be one of the potential solutions which could be used for fueling diesel engines [6-8].

Developing precise and robust models to describe the properties of newly developed fuels for ICE is compulsory not only in order to predict all parameters of the production, storage and transportation of the fuel [9], but also to describe thermodynamic processes related to internal combustion engines [10].

Density is one of the most important properties of the fuels because it influences its production, transportation and distribution processes, and all the processes which occur in an ICE [11]. Apart from viscosity, surface tension, and vapor pressure, density determines the fuel spray quality and thus the combustion and emission characteristics of the engine [12]. Fuel density is the main property that influences the amount of injected fuel mass and therefore, the fuel consumption [11].

Viscosity affects the quality of the fuel spray formation, the fuel droplet size, the penetration of the fuel jet and,

ultimately, the quality of the combustion which influences the efficiency and emission level of an ICE [13]. Fuel viscosity influences the necessary pumping power, fuel leakage losses in the fuel pump and the fuel lubricity [14].

Ethanol has a very limited miscibility in a diesel fuel. Using proper surfactants, like methyl esters or long-chain alcohols, their miscibility can be improved [15].

THF was identified as an effective co-solvent for diesel-ethanol blends and it can be obtained at low cost from agricultural waste materials [16]. Research has pointed out that blending THF with other fuels, like algae biodiesel [17] or soybean oil [18], reduces the emissions from the diesel engine.

There are some concerns about the reduction of the flash point and the cetane number of the blends owing to the low flashpoint and cetane number of ethanol and THF [19]. Whereas the flash point of the blend cannot be easily increased, the cetane number can be restored by using a small amount of cetane improver additive [20].

Including a third component into the diesel fuel – ethanol blends makes the classical modeling methods more difficult. The density and the viscosity of diesel fuel – ethanol blends were investigated in many studies but, to the best of our knowledge, models for densities and viscosities of diesel fuels – ethanol – surfactant blends, which take into account, in the same time, both the composition and the temperature of the blends, have not been studied yet.

The main aim of this work is to develop proper mathematical models which are able to accurately describe both the temperature and composition dependences of densities and absolute (dynamic) viscosities of the diesel fuel – ethanol blends stabilized by a surfactant.

2 Materials and methods

In this research commercially available diesel fuel, ethanol with 99.2 % purity and tetrahydrofuran (THF) having 99.5 % concentration were used. The diesel fuel used in this research followed the European standard EN 590 for winter fuel and its main properties related of this work were evaluated using an IROX Diesel type apparatus. Its aromatic hydrocarbon content was 42.9 % (v/v), whereby 7.4 % (v/v) was polyaromatic hydrocarbon and it included 0.2 % (v/v) biodiesel.

Initially, using class A precision pipettes and graded cylinders, five diesel fuel – ethanol blends were prepared at room temperature (20 °C), containing 2, 5, 8, 10 and 15 % (v/v) ethanol. These blends were homogenized in a closed Berzelius beaker with a magnetic stirrer

at 400 r/min for 30 minutes and in an ultrasonic bath for a further 20 minutes. After the preparation, the blends were monitored for three days in order to evaluate their stability and miscibility. In the case of the blends which presented any phase separation, depending on the quantity of separated ethanol, 0.5 mL, 1.0 mL or 2.0 mL of tetrahydrofuran were added as a surfactant into the blends. These new blends were homogenized and monitored again in the same way as aforementioned. These steps were repeated until all the blends became stable. At this point, the compositions of the blends were recalculated and expressed in relative volume of diesel fuel, ethanol and tetrahydrofuran.

The temperature dependent densities and viscosities of the components and the blends were measured by using an SVM 3000 type apparatus. It measured the absolute viscosity and density (ASTM D7042 [21] and ISO 12185 [22]) and calculated the kinematic viscosity. The uncertainties of the density and viscosity measurements at the 95 % confidence level with a coverage factor of $k = 2$ were estimated 0.02 % for density and 0.35 % for viscosity.

Prior to the measurements, the viscometer was cleaned with a proper solvent, it was recalibrated with standardized fluids and finally it was dried with air. All the measurements were repeated three times and their average values were considered. Because of the high volatility of the THF (its boiling point is 64 °C) the viscosity and density measurements were carried out up to 50 °C at four temperatures: 0 °C – which evaluates the cold flow properties, 15 °C – the standard temperature for density measurement, 40 °C – the standard temperature for viscosity measurement) and 50 °C – the maximum temperature for accurate measurements.

The measured data were processed, and the mathematical models were developed with the aid of ORIGIN software, in the case of the components, and EXCEL-Solver software by minimizing the Average Relative Deviation of the models, in the case of the blends.

The accuracy of the developed models was evaluated based on their Average Relative Deviation (ARD), Standard Deviation (SD), and correlation coefficient (R) which are defined by the following equations:

$$ARD = \frac{100}{N} \sum_{i=1}^N \frac{|est_i - exptl_i|}{exptl_i} \% \quad (1)$$

$$SD = \sqrt{\frac{\sum_{i=1}^N (est_i - exptl_i)^2}{N - m}} \quad (2)$$

$$R = \frac{\sum_{i=1}^N (\overline{exptl}_i - \overline{exptl}) (\overline{est}_i - \overline{est})}{\sqrt{\sum_{i=1}^N (\overline{exptl}_i - \overline{exptl})^2 \sum_{i=1}^N (\overline{est}_i - \overline{est})^2}} \quad (3)$$

where \overline{exptl}_i and \overline{est}_i are the measured and estimated values, \overline{exptl} and \overline{est} are the averages of the measured and predicted values, N represents the number of experimental values and m is the number of model parameters.

3 Experimental results

After the prepared blends had become stable, their composition was changed because of the THF addition. The final compositions of the stabilized blends are presented in Table 1. The average ratio of THF to ethanol to form stable blends at room temperature was 1:2.2.

The measured temperature-dependent densities and viscosities of the blends are given in Table 2. The maximum Standard Deviations of the experimental measurements were 0.0001 in the case of the density and 0.028 in the case of absolute viscosity.

Table 1 The composition of the stabilized diesel – ethanol – THF blends

Components	Initial concentration, % (v/v)				
	2	5	8	10	15
Ethanol	2	5	8	10	15
	Concentrations after stabilization, % (v/v)				
Diesel	97.512	92.683	87.559	84.791	79.017
Ethanol	1.990	4.878	7.614	9.421	13.944
THF	0.498	2.439	4.827	5.787	7.039

Table 2 The measured densities and viscosities of diesel – ethanol – THF blends

Temp., °C	Components			Initial concentration, % (v/v)				
	Diesel	Ethanol	THF	2	5	8	10	15
Density, ρ , g/cm ³								
0	0.8522	0.8160	0.9100	0.8393	0.8385	0.8391	0.8393	0.8388
15	0.8416	0.8034	0.8937	0.8286	0.8273	0.8278	0.8278	0.8270
40	0.8240	0.7816	0.8661	0.8109	0.8088	0.8087	0.8085	0.8073
50	0.8170	0.7728	0.8535	0.8038	0.8013	0.8011	0.8007	0.7994
Absolute viscosity, η , mPa·s								
0	6.7785	2.0847	0.6102	5.2687	4.5151	4.1170	3.9289	3.8771
15	4.2290	1.4901	0.5389	3.3658	2.9069	2.6540	2.5221	2.4241
40	2.2687	0.9061	0.4381	1.9244	1.6601	1.5422	1.4705	1.3857
50	1.8587	0.7121	0.4041	1.6032	1.3730	1.2859	1.2334	1.1564
Kinematic viscosity, ν , mm ² /s								
0	7.9540	2.5547	0.6705	6.2778	5.3851	4.9064	4.6814	4.6221
15	5.0247	1.8548	0.6030	4.0618	3.5135	3.2060	3.0467	2.9311
40	2.7532	1.1593	0.5058	2.3733	2.0527	1.9069	1.8187	1.7164
50	2.2750	0.9215	0.4734	1.9945	1.7135	1.6053	1.5404	1.4466

As the density of THF is higher than the density of the other two components it compensates the lower density of the ethanol. Consequently, the density of the blends fulfills the EN 590 standard requirement (0.820-0.845 g/mL at 15 °C). In the case of viscosity, only the blends containing up to 5 % (v/v) ethanol meet the viscosity requirement (2.0-4.5 mm²/s at 40 °C) for diesel fuel; for the rest of the blends it falls below 2.0 mm²/s. The maximum decrease is 14 % and it belongs to the blend with 15 % ethanol.

4 Models for fuel blends properties

4.1 Models for predicting the densities of the blends

In our previous work [23] we concluded that a quadratic model was a good estimator for modeling the temperature-dependent density of the components:

$$\rho(t) = a_0 + a_1 t + a_2 t^2, \quad (4)$$

where $\rho(t)$ (g/mL) is the density, t (°C) is the temperature, a_0 , a_1 and a_2 are parameters of the regressions. The calculated values of the regression parameter for each compound and the precision of the models are presented in Table 3.

It can be observed that all the models describe the temperature-dependent densities of the components with a very high fidelity, having the regression coefficient almost 1 and a very low Average Relative Deviation (below 0.5 %).

One of the widely used methods to estimate properties of liquid blends is based on the mixing of the properties of its components by using a suitable mixing rule.

Table 3 Parameters and accuracies of density models of the components

Parameters	Diesel	Ethanol	Tetrahydrofuran
a_0	0.8522	0.8160	0.90987
a_1	$7.0827 \cdot 10^{-4}$	$-8.3518 \cdot 10^{-4}$	-0.0010
a_2	$8.3596 \cdot 10^{-8}$	$-5.9685 \cdot 10^{-7}$	$-1.8638 \cdot 10^{-6}$
R	0.9999	0.9999	0.9999
SD, g/mL	$3.7009 \cdot 10^{-6}$	$4.0200 \cdot 10^{-5}$	0.0003
ARD, %	0.0004	0.0048	0.0300

Kay's Mixing Rule describes the properties of blends by the concentration-weighted sum of the properties of the components. For density, the Kay's Mixing Rule (KMR) is [11, 24]:

$$\rho_{est} = f_D \rho_D(t) + f_E \rho_E(t) + f_T \rho_T(t), \quad (5)$$

where $\rho_{est}(t)$ (g/mL) is the predicted density of the blend, $\rho_D(t)$, $\rho_E(t)$ and $\rho_T(t)$ (g/mL) are the temperature dependent densities of diesel, ethanol and THF, given by Eq. (4), f_D , f_E and f_T are the volume fraction of the diesel, ethanol and THF.

Since Eq. (5) has a positive error of 1.6 % (Fig. 1 a), Table 4), its precision must be improved. In order to fine

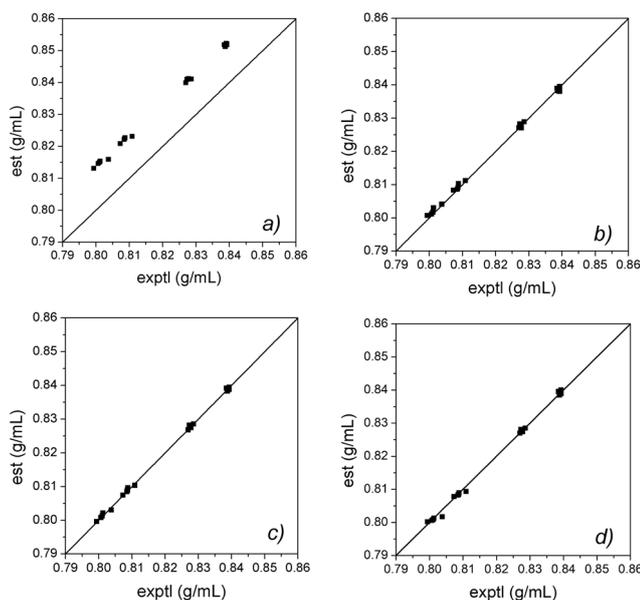


Fig. 1 The precisions of the densities models:
a) Eq. (5); b) Eq. (6); c) Eq. (7); d) Eq. (9).

tune the model, the contributions of the components to the density of the blend have to be weighted [24]:

$$\rho_{est}(t) = k_D f_D \rho_D(t) + k_E f_E \rho_E(t) + k_T f_T \rho_T(t), \quad (6)$$

where k_D , k_E and k_T are weighting parameters. This model improves the density prediction accuracy, having an ARD of 0.0826 (Fig. 1 b) and Table 4).

The precision of Eq. (6) can be further improved by introducing a constant term, k_0 :

$$\rho_{est}(t) = k_D f_D \rho_D(t) + k_E f_E \rho_E(t) + k_T f_T \rho_T(t) + k_0. \quad (7)$$

This proposed model halves the ARD of the previous model (Fig. 1 c), Table 4). As the influence of the temperature is a linear one, it can be modeled as monomial:

$$\rho_{est}(t) = k_D f_D \rho_D(t) + k_E f_E \rho_E(t) + k_T f_T \rho_T(t) + k_1 t + k_0, \quad (8)$$

where k_1 and k_0 are constants. The precision of this model is comparable with the precision of Eq. (7).

Usually, only the density at 15 °C of the diesel fuel is known and the densities of the other two components are material constants. In this case the temperature- and composition – dependent densities of the blends can be estimated using the following model:

$$\rho_{est}(t) = k_D f_D \rho_D^{15} + k_E f_E \rho_E^{15} + k_T f_T \rho_T^{15} + k_1 t + k_0, \quad (9)$$

where ρ_D^{15} , ρ_E^{15} and ρ_T^{15} are the densities of diesel fuel, ethanol and THF respectively. As it is shown in Fig. 1 d) and Table 4, the precision of this model is lower than the precision of the previous two models, but it has the important advantage of using only one experimental value (the density of the diesel fuel at 15 °C).

4.2 Models for predicting the viscosities of the blends

The temperature dependence of the absolute viscosity of the components can be modeled by the Vogel-Fulcher-Tammann (VFT) equation [25]:

$$\ln[\eta(t)] = A + B/(C + t), \quad (10)$$

Table 4 The constants and the accuracies of the models described by Eqs. (5)-(9)

Eq.	k_D	k_E	k_T	k_1	k_0	ARD, %	SD, g/mL	R
(5)	–	–	–	–	–	1.6163	0.0162	0.9993
(6)	0.9849	1.0526	0.8618	–	–	0.0826	0.0147	0.9992
(7)	0.4125	0.0081	0.0022	–	0.4964	0.0444	0.0149	0.9995
(8)	0.01634	0.0078	0.0023	-0.0008	0.8264	0.0595	0.0151	0.9999
(9)	0.0157	0.0082	0.0023	-0.0008	0.8270	0.0604	0.0151	0.9989

where $\eta(t)$ (mPa·s) is the absolute viscosity of the component at temperature t (°C) and A , B and C are parameters which can be calculated by measuring the viscosity at least at three different temperatures. The values of the regression parameters and the precision of the models are given in Table 5.

As in the case of the densities, the absolute viscosities of the blends can be described using Kay's Mixing Rule which is a simplified Grunberg-Nissan model [25, 26]:

$$\ln[\eta_{est}(t)] = f_D \ln[\eta_D(t)] + f_E \ln[\eta_E(t)] + f_T \ln[\eta_T(t)], \quad (11)$$

where $\eta_{est}(t)$ (mPa·s) is the predicted viscosity of the blend, $\eta_D(t)$, $\eta_E(t)$ and $\eta_T(t)$, (mPa·s) are the temperature dependent viscosities of diesel, ethanol and THF, given by Eq. (10), f_D , f_E and f_T are the volume fraction of the diesel, ethanol and THF respectively.

As shown in Fig. 2 a) and Table 6, Eq. (11) has a large positive error of 27 %. Das et al. [27] proposed an improved

model for the viscosity of a ternary blend by adding a fractional term to Eq. (11):

$$\ln[\eta_{est}(t)] = f_D \ln[\eta_D(t)] + f_E \ln[\eta_E(t)] + f_T \ln[\eta_T(t)] + \frac{M_0}{t + M_1}, \quad (12)$$

where M_0 and M_1 are constants. By minimizing the Average Relative Deviation (ARD) of the prediction model, the resulted values of these constants are $M_0 = -52.0906$ and $M_1 = -179.5159$. This model improves the precision of the prediction, mainly the ARD which drops more than three times (Fig. 2 b), Table 6).

Another way to improve the precision of Eq. (11) is proposed in [11, 24] by using weighting factors:

$$\ln[\eta_{est}(t)] = k_D f_D \ln[\eta_D(t)] + k_E f_E \ln[\eta_E(t)] + k_T f_T \ln[\eta_T(t)], \quad (13)$$

where all the notations have the same meaning as defined above. As illustrated in Fig. 2 c) and evaluated in Table 6, the precision of this model is good for the proposed applications.

Usually, the viscosity of the diesel fuel is given at the temperature of 40 °C. Therefore, a model which only uses the viscosities of the components at this standardized temperature can be more useful in most of the practical applications:

$$\ln[\eta_{est}(t)] = k_D f_D \ln(\eta_D^{40}) + k_E f_E \ln(\eta_E^{40}) + k_T f_T \ln(\eta_T^{40}) + k_0 t, \quad (14)$$

where η_D^{40} , η_E^{40} and η_T^{40} are the absolute viscosities of the components at 40 °C, all other notations have the same meaning as they were defined above. This model delivers a lower precision than Eq. (13) (Fig. 2 d), Table 6), but it is very practical.

The kinematic viscosity of the blends can be calculated as a ratio of absolute viscosity to density. Using Eq. (13) for the absolute viscosity and Eq. (8) for the density, the resulted values of the kinematic viscosity are in very good agreement with the experimental values (ARD = 2.2579 %, Fig. 3 a). Even when the less precise models (Eqs. (9) and (14)) are used, the precision of the resulted kinematic viscosities remains acceptable (ARD = 3.6813 %, Fig. 3 b)).

4.3 Validation of the developed models for other blends
 All the constants of the developed models were determined for the diesel fuel, ethanol and THF used in this work. In order to validate the models, they were tested with viscosity and density data obtained from the existing

Table 5 Parameters and accuracies of viscosity models of the components

Parameters	Diesel	Ethanol	Tetrahydrofuran
A	-3.2261	-7.0357	-6.3491
B	762.0029	2510.4069	3874.8473
C	148.2529	322.9650	661.5741
R	0.9999	0.9999	0.9999
SD, mPa·s	$7.3636 \cdot 10^{-3}$	$33.2516 \cdot 10^{-3}$	$2.5350 \cdot 10^{-3}$
ARD, %	0.0004	0.0048	0.0300

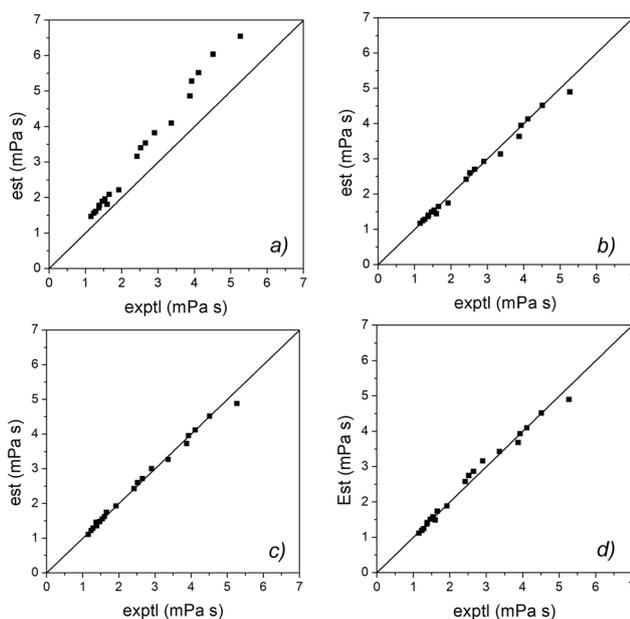
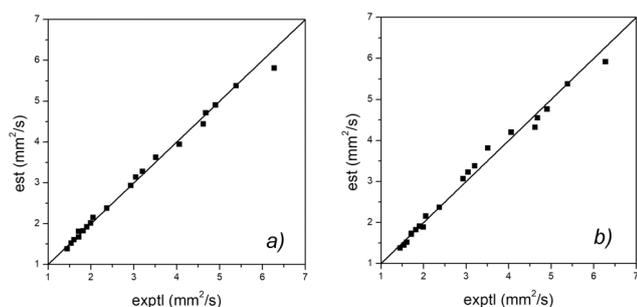


Fig. 2 The precisions of the viscosity models: a) Eq. (11); b) Eq. (12); c) Eq. (13); d) Eq. (14).

Table 6 The constants and the precision of the models described by Eqs. (11)-(14)

Eq.	k_D	k_E	k_T	k_0	ARD, %	SD, mPa·s	R
(11)	–	–	–	–	27.1054	1.4859	0.9958
(12)	–	–	–	–	7.8774	1.2105	0.9961
(13)	0.8412	1.6846	3.7659	–	2.2208	1.2086	0.9972
(14)	1.9665	–9.7039	2.3180	–0.02387	3.8931	1.2166	0.9939

**Fig. 3** The precisions of the predicted kinematic viscosity:
a) Eq. (13)/Eq. (8); b) Eq. (14)/Eq. (9).

literature. Unfortunately, relevant literature provides very few data on the density and viscosity of diesel-ethanol-tetrahydrofuran blends. Moreover, these data are published for one single value of the temperature and not for an interval, as it is the case of the present study.

The validation of the density models based on data from [28] is presented in Table 7. It can be observed that the density models developed in this research provide a very good precision. The model given by Eq. (8) has the best precision, followed by the models described by Eq. (9) and Eq. (7).

In terms of viscosity, using the data published in the same source [28], the models for absolute viscosity have been verified, and the results are presented in Table 8. It can be noticed that the viscosity models developed in the present study (Eqs. (13) and (14)) deliver a good accuracy

in the case of other types of blends, better than models from the specialized literature (Eqs. (11) and (12)).

5 Conclusions

The temperature- and composition – dependent density and viscosity of diesel fuel – ethanol blends containing up to 15 % (v/v) ethanol were modeled in the temperature range of 0-50 °C. Based on our findings, the following conclusions can be framed:

- Blends containing more than 2 % ethanol are not stable at room temperature, they start separating within a few minutes after the homogenization process stops. Above this concentration they require a surfactant, like tetrahydrofuran. The necessary quantity of surfactant increases as the ethanol quantity increases in the blend.
- In the case of the density of the blends, the weighted Kay's rule is in good agreement with the experimental data.
- The weighted Key's rule, completed by a monomial, delivers a very good prediction of the temperature and composition dependent viscosity.
- Given that density and viscosity are material properties in the case of ethanol and tetrahydrofuran, the models described by Eqs. (9) and (14) are able to describe these quantities as a function of their temperature and composition using a single

Table 7 Density models validation

Components			Density at 20 °C, g/cm ³			
			Measured	Calculated		
Diesel	Ethanol	THF		Eq. (7)	Eq. (8)	Eq. (9)
97.4	2.0	0.6	0.8250	0.8332	0.8239	0.8240
95.0	4.0	1.0	0.8257	0.8251	0.8237	0.8238
92.0	6.0	2.0	0.8260	0.8148	0.8234	0.8236
87.6	8.0	4.4	0.8263	0.7998	0.8230	0.8232
85.0	10.0	5.0	0.8243	0.7910	0.8228	0.8230
80.0	15.0	5.0	0.8208	0.7740	0.8224	0.8227
ARD, %				2.5622	$2.03 \cdot 10^{-5}$	0.0023
SD				0.0178	0.0016	0.0016
R				0.9059	0.9803	0.8754

Table 8 Viscosity models validation

Components			Absolute viscosity at 40 °C, mPa·s				
Diesel	Ethanol	THF	Measured	Calculated			
				Eq. (11)	Eq. (12)	Eq. (13)	Eq. (14)
97.4	2.0	0.6	2.1179	2.2056	2.1942	1.9141	1.9536
95.0	4.0	1.0	1.9865	2.1513	1.3734	1.8532	1.9012
92.0	6.0	2.0	1.8549	2.0777	1.3875	1.7539	1.8116
87.6	8.0	4.4	1.7868	1.9609	1.3734	1.5740	1.6430
85.0	10.0	5.0	1.6581	1.9064	1.3734	1.5125	1.5877
80.0	15.0	5.0	1.5772	1.8209	1.3734	1.4491	1.5366
ARD, %				10.7676	18.8165	8.4319	4.8758
SD				0.1961	0.3110	0.2039	0.1856
R				0.9838	0.7023	0.9757	0.9729

experimental value (the density of the diesel fuel at 15 °C in the case of Eq. (9) and its viscosity at 40 °C in the case of Eq. (14), respectively).

- Calculating the kinematic viscosity by using the proposed density and viscosity models delivers very precise results in good agreement with the experimental values.
- The density of the blends fulfills the EN 590 standard requirement, but only blends with up to 5 % ethanol meet the viscosity limits imposed by the same standard.

- The developed models can be used in the proposed applications.

The authors intend to develop further models to describe other properties of diesel-ethanol-THF blends in order to use them in fuel injection and combustion modeling and simulation. Furthermore, preliminary internal combustion engine tests have been already done with promising results.

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