Нові рішення у сучасній техніці та технологіях

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MIXTURES OF DIETHYL ETHER AND LINSEED OIL AS AN ALTERNATIVE FUEL FOR DIESEL ENGINES

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Фізичні та хімічні властивості паливних сумішей діетилового ефіру та лляної олії (DEE/LO) були вперше емпірично перевірені в цій статті. Зокрема, досліджували кінематичну в'язкість, щільність, нижню теплотворну здатність, точку закупорювання холодного фільтра та поверхневий натяг. Також були запропоновані моделі прогнозування для згаданих емпіричних даних. Для цього дослідження діетиловий ефір (DEE) змішували з лляною олією (LO) в об'ємних співвідношеннях 10, 20 і 30%. Отримані результати порівнювали з літературними даними про паливо з діетилового ефіру рапсової олії (DEE/RO). Ці тести були зосереджені на основних параметрах роботи двигуна, таких як загальна ефективність і затримка запалювання. Таким чином, процес згоряння є більш ефективним – з меншою кількістю викидних газів. Відповідно до різних застосувань рослинні олії можуть бути успішно використані в двигунах у складі класичних палив та модифікованих двигунах. Як альтернативним паливам, все більшу увагу приділяють неїстівним рослинним оліям, що пов'язано з виснаженням природних ресурсів і потребою в харчових оліях як продуктах харчування. Насіння льону практично використовується в промислових цілях замість найпопулярнішої ріпакової олії, але, виходячи з його технічних характеристик, за певних умов його можна використовувати і як паливо. Результати цього дослідження показали, що в'язкість LO значно нижча порівняно з RO, що дозволяє зменшити кількість DEE, доданого до рослинної олії. Наприклад LO, що містить 30% DEE, має таку саму в'язкість, як і RO, що містить 40% DEE. Слід зазначити, що обидві ці суміші дозволяють збільшити значення в'язкості в порівнянні з DF. Результати підтвердили, що кінематична в'язкість досліджуваних сумішей виражається експоненціальною регресією на основі рівняння Френкеля-Андраде.

Ключові слова: діетиловий ефір, рослинні олії, паливні суміші, властивості палива, альтернативне паливо, рівняння Френкеля-Андраде.

The physical and chemical properties of diethyl ether and linseed oil (DEE/LO) fuel mixtures were empirically tested in this article for the first time. In particular, kinematic viscosity, density, lower heating value, cold filter plugging point, and surface tension were examined. Prediction models for the mentioned empirical data were also proposed. For this research diethyl ether (DEE) was blended with linseed oil (LO) in volumetric ratios of 10, 20 and 30%. The results obtained were compared with the literature on diethyl ether rapeseed oil (DEE/RO) fuel. These tests were focused on the fundamental engine work parameters such as overall efficiency, and ignition delay. Therefore, the combustion process is more efficient with adequate lower exhaust gases. According to different applications, vegetable oils can be successfully used in engines based on classic fuels and modified engines. More and more attention is turned on inedible vegetable oils affected by the depletion of resources and the necessity for edible oils as food. Linseed is practically used for industrial purposes instead of the most popular rapeseed oil, but based on its technical characteristics it can be used also as a fuel under certain conditions. Results of this study showed that the *viscosity of LO is significantly lower in comparison with RO allowing to reduced amount of DEE added to plant oil. For example, LO containing 30% DEE has the same viscosity as RO containing 40% DEE. It should be pointed out that both these mixtures allow to increase a viscosity value in comparison to DF. The results confirmed that the kinematic viscosity of tested mixtures is expressed by exponential regression based on the Frenkel-Andrade equation.*

Keywords: diethyl ether, plant oils, fuel mixtures, fuel properties, alternative fuel, the Frenkel-Andrade equation.

Introduction

Vegetable oil as a diesel engine fuel has been a subject of study for a long time, probably from the first usage of it in Rudolf Diesel's invented engine. Although vegetable oils did not become as important a fuel source as fossil fuel as Diesel predicted during the exploitation of his engine with peanut oil, there were and still exist some areas where vegetable oils are requested and used. The usage of vegetable oils is usually outpointed by its main advantages over conventional diesel fuels: domestic production allows to reduce dependency on imported diesel fuel, renewability, better lubricity, biodegradability, and increase of flash point. As with any other alternative fuel, it could outpoint also some disadvantages, which did not allow its rapid usage during the last decade: lower energy content, higher viscosity, compatibility, higher production price, etc. Despite to different application problems vegetable oils can be successfully used in engines based on fuel or engine modification. More and more attention is turned on inedible vegetable oils affected by the depletion of resources and the necessity for edible oils as food. Linseed instead of the most popular rapeseed oil is practically used for industrial purposes, but based on its technical characteristics it can be used also as a fuel under certain conditions.

Analysis of modern foreign and domestic research and publications

Linseed is recognized as one of the oil crops good for the production of advanced biofuels by the European Biofuels Technology Platform (EBTP), but it is also used as a first-generation biofuel obtained from seeds. Linseed oil is very valuable and one of the oldest commercial oils produced from the oilseed flax *(Linum usitatissimum L.)* widely grown in Eastern Europe, Kazakhstan, India, and China. Due to its low-level yield, it is not as popular as soybean or rapeseed the leading oilseed crops, which provide the largest amounts of plant protein in an extracted meal for livestock [1] but in some countries like Kazakhstan cultivation areas of this plant is growing very rapidly allowing to become the world's undoubted leader in linseed production with harvested 719 km in 2018. About 70% of worldwide linseed oil production is intended for technical applications and only 30% for food production [2]. Many different parameters, like water stress, high temperature, and disease occurrence, can affect oilseed flax growth parameters [3], but increasing demand from the paints and coatings industry, where linseed oil is very popular as a varnish and as a drying lubricant, it is expected to boost the market over the forecast period. Increasing applications of linseed oil in food processing, paint, wood finish, putty, nutritional supplement, linoleum, and gilding are expected to boost the segment growth, which be valued at USD 281.34 million by 2025 [4].

There are not many practical researches or on-road trials concerning linseed oil due to previously mentioned factors, while there are also some researches for oils with similar composition allowing to make general conclusions. For example, Beg et al. [5] reported an increase in fuel consumption, exhaust gas temperature, CO emissions, smoke density, and a decrease in NOx using diesel fuel-linseed blends compared to regular diesel fuel. Delalibera et al. [6] analysed diesel engine performance and emissions when fuelled with pure linseed oil, preheated at 100 °C, at the injection pump inlet, and at engine working temperature (60 °C) in short-duration tests. Results confirmed a slight increase in exhaust gas temperature generally for preheated oil and a slight reduction of smoke for non-preheated oil compared to diesel fuel, while smoke increased for preheated oil compared to diesel fuel. Fuel consumption for preheated oil presented lower variation when compared to non-preheated oil indicating that the engine operation for them was more stable. He confirmed that an engine without linseed oil preheating presents greater power loss compared to diesel fuel at the same time.

Agarwal et al. [7] tested different blends of linseed oil (10, 20, 30, and 50%, v/v) with diesel fuel and found that blends with higher linseed oil addition are more efficient. Overall it is concluded that vegetable oils are not ideal fuels for diesel engines in case of operational (ignition, performance, etc.) and durability (deposit formation, lubrication oil dilution, etc.) problems, but also based on additional impact left by other physicochemical properties: viscosity, polyunsaturated character, and extremely low volatility. Based on that diesel engine operation with vegetable oil could result in poor atomization, non-ideal fuel-air mixing and combustion, and finally in decrease in power and an increase in fuel consumption. Different solutions exist to overcome such problems: transesterification, emulsification, and blending. One of the most popular is transesterification allowing to convert vegetable oil in an ester in such way creating fuel with properties close to diesel fuel. Another, more simplified, and attractive alternative is blending vegetable oil with diesel fuel or oxygenate, which drastically reduces viscosity, and increases cold filter plugging point and cetane number. One of the main blending problems is observed – its molecular structure remains unchanged hence polyunsaturated character remains [8].

Improvement of oil properties could be realized using different oxygenated additives such as ethanol, methanol, ethyl-test butyl ether, diethyl ether (DEE), etc. The last one was chosen for this research. Diethyl ether, which is an organic compound and historically used as an anesthetic agent is expressed by its chemical formula CH3CH2–O–CH2CH3. Produced from ethanol obtained from biomass in the dehydrating process, it also can be known as bio–DEE [8], showing its main advantages over diesel fuel: high oxygen content, high cetane number, high miscibility with diesel fuel, and low autoignition temperature. First of all, this type of fuel stands up with low viscosity values allowing to reduce the viscosity of the blend, whereas base fuel has a viscosity higher than that of conventional diesel fuel, like plant oils. The combination of these values of both mentioned fuel types could give the optimal viscosity necessary for diesel engine operation without preheating and at the same time keeping lubricity at reasonable levels.

Rakopoulos [9] used a standard, experimental, single-cylinder, four-stroke, high-speed direct injection (HSDI) diesel powered with cottonseed oil and its bio-diesel (methyl ester) in blends with 20% (v/v) of diethyl ether for research of combustion and exhaust emission characteristics. He reported lower smoke, NOx, and CO emissions, but higher HC emissions for DEE blends than neat cottonseed oil or its neat bio-diesel.

Krishna [10] used diethyl ether as an oxygenated additive mixed with karanja oil in various proportions of 5, 10, 15, 20, and 25% by volume in dual fuel operation 4-cylinder 39 kW diesel engine. He observed that for blends up to 15%, CO emissions were lower than corresponding values for diesel, while NOx emissions were significantly lower for the blends at lower loads. He also found that fuel consumption at a higher engine loads for the blend is higher than for pure diesel explaining it with the lower calorific value of the blends.

Geo [11] used a single-cylinder diesel engine with a rated output of 4.4 kW at 1500 rpm converted for operation in the DEE injection mode. The engine was operated on rubber seed oil as a single fuel with DEE injection at the flow rate of 100 g/h, 150 g/h, and 200 g/h. He reported an increase in NOx emissions and a reduction of hydrocarbon, carbon monoxide, and smoke emissions, the last one explaining with "better combustion of injected fuel in the hotter combustion chamber by the early combustion of DEE". Besides that, he confirmed that DEE injection to rubber seed oil results in reduced combustion duration from 47 to 44 crank angle (°CA) degrees due to fast diffusion combustion rate in comparison to neat oil.

Krishnamoorthi [12] used a single-cylinder direct injection variable compression ratio test engine Kirloskar VCR in tests with blends of diesel fuel, bael oil, and diethyl ether in various blending ratios. They confirmed that the addition of DEE to bael oil and diesel reduced the peak cylinder pressure due to a lower calorific value of the DEE and wider spray pattern. It was also observed CO emission reduction explained with enhancement of the combustion process with the effect of DEE. There was also a reported reduction of NOx emissions associated with an adequately lower value of peak combustion temperature due to DEE addition in such a way as reducing the calorific value of the blend.

There is no research on detailed analysis of the physicochemical and combustion properties of linseed oil and DEE blends, while there exists research on the physic chemicals of rapeseed oil and DEE blends [13]. Therefore, the objective of this study was to test the effect of DEE/linseed oil fuel blends to evaluate the possibility of using such mixtures in diesel engines, as well as make a comparison of obtained results with DEE/RO blends.

Highlighting previously unresolved parts of the overall problem

Vegetable oils containing high polyunsaturated fatty acids are less viscous but contribute to higher NOx emissions. Therefore, the usage of vegetable oils with higher saturated and monounsaturated fats together with the retrofit kit or heating system was recommended. High values of linoleic/linolenic acids also could result in higher deposition rates as these acids react to heat and oxidize polymerizing on the cylinder walls and injection tips.

Possible linseed oil usage in diesel engines is strongly connected to the engine sensitivity on fuel injection and combustion, and one of the main parameters is viscosity. Viscosity which is a measure of the resistance of fluid to a flow, is the most important parameter for all vegetable oils as it leaves an impact on the quality of fuel atomization. The fuel injection system of current diesel engines accepts the use of fuels with viscosity values from 1.9 to 5.0 mm^2/s at 40 °C (1.9 to 4.1 mm2/s at 40 °C based on standard ASTM D975, 2.0 to 4.5 mm^2/s at 40 °C from standard EN 590:2004, 3.5 to 5.0 at 40 °C from EN 14214:2009.

Cetane number is a dimensionless descriptor of the ignition quality of diesel fuel, determined by unbranched chains of fatty acids similar to those of the n-alkanes of diesel fuel. Chain length leaves an impact on cetane number, where decreasing chain length reduces cetane number. There could be determined also a relationship between cetane number and ignition delay, where the higher cetane number determines a shorter ignition delay. As previously it was mentioned – linseed oil has one of the longest ignition delays among vegetable oils and one of the smallest cetane numbers – 34.6 instead of 37.6 for rapeseed oil or 42.0 for palm oil. Therefore, the influence of increased unsaturation on the lowest cetane number is proven for linseed oil. An increase in cetane number is possible by blending oil with alcohols (like DEE), while the heating value of the blends will be practically in the same range.

As mentioned before, the role of viscosity in the injection process is more actual than for any other parameter, therefore it must be set according to values mentioned in standards: EN 590, ASTM D975, or EN 14214. Spray characteristics significantly can be improved with the use of blends compared to neat linseed oil. The viscosity of LO is lower than for RO allowing it to reach more acceptable values for diesel engines even with a small addition of DEE, till 30%. For blends with DEE addition of 40% and more in volume variations in viscosity are not so actual and are not influenced by oil type. This confirms the efficiency of blending compared to preheating and engine conversion for oil use.

The purpose and tasks of research

To study the main characteristics (density, viscosity, surface tension, CFPP with respect to ratio of DEE/LO, and lower heating value) of diethyl ether and linseed oil mixture as an alternative fuel for diesel engines.

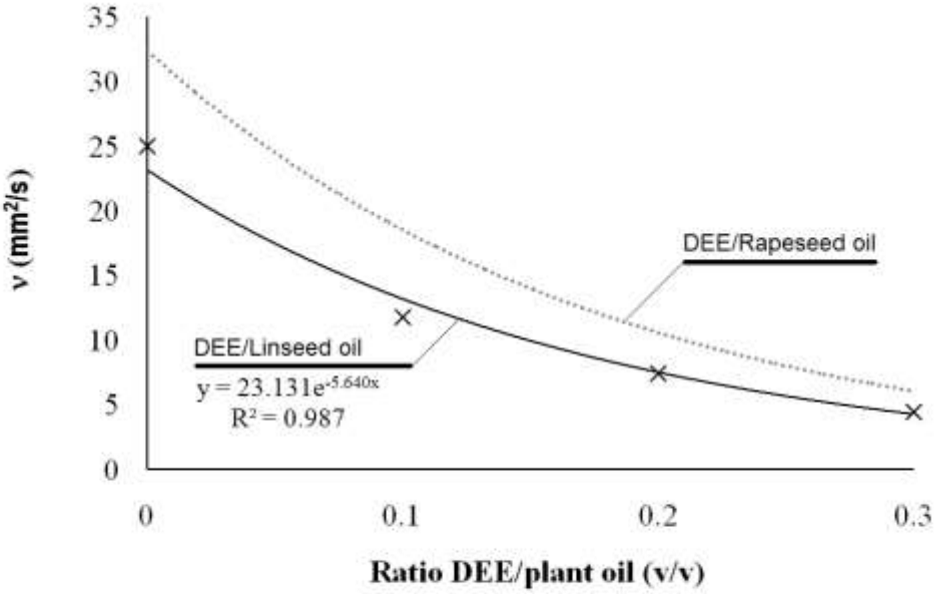
Highlighting of the main research material

Parametric regression parameters for obtained experimental dependence for kinematic viscosity ν calculated by the least squares method are expressed as $\nu = y = 23.131e^{-5.640x}$. However, ν is described by Frenkel-Andrade exponential law more precisely i.e. $\nu = Ce^{\frac{E_a}{kT}}$, where E_a denotes activation energy, k is Boltzmann constant, which describes kinetic theory of a fluid by finding kinematic viscosity and Arrhenius equation. Cofactor C characterizes temperature, length of jump of molecule and frequency of its oscillation, although influence of temperature on viscosity is mainly determined by component $e^{\frac{E_a}{kT}}$ and with increasing of temperature viscosity of liquids decreases rapidly.

Viscosity of fluid in experiments is determined by forces of intermolecular interaction with each other. In this case thermal motion of molecules is reduced mainly to oscillations inside potential trough. But if due to thermal fluctuation molecule receives energy greater than depth of potential trough, it can jump to the next free space formed by leaving the other molecule. Minimum energy required to jump a molecule is called activation energy E_a.

Most of liquid molecules are situated near equilibrium positions therefore momentum transfer due to spontaneous jumps of molecules from layer to layer as in gas is very small. Moving mass of fluid attracts the adjacent layers mainly due to the forces of molecular adhesion. Viscosity of a fluid is thus determined by number of molecules that have received energy greater than activation energy sufficient to jump into direction of the layers' motion under action of adhesion forces, and concentration of nearby vacancies where the molecule can jump. Both of these quantities according to the Boltzmann distribution must be proportional to $e^{-\frac{E_a}{kT}}$ therefore fluidity increases with increasing of temperature. It follows that viscosity of liquid is proportional to $e^{-\frac{E_a}{kT}}$ and it decreases with increasing temperature.

Modern automobiles must be operated in a wide range of ambient temperatures. Moreover, in modern diesel engines fuel is compressed even up to 200 MPa or more and as a result it leads to significant heating of diesel fuel. In addition, it should be noted that increasing of pressure increases activation energy and reduces concentration of vacancies, therefore at high pressures viscosity of liquids increases. Therefore, finding respecting between



LO – continuous line; RO – dashed line

Figure 1 – Kinematic viscosity (v) of DEE/LO and DEE/RO blend at 40 °C

viscosity and temperature of diesel fuel is an important practical challenge to ensure efficient fuel injection.

Obtaining value of activation energy, Andrade coefficients at different temperatures and ratio of liquids DEE/LO are expressed from the following eq:

$$C = \nu e^{-\frac{E_a}{kT}} . \qquad (1)$$

Dependence of reaction rate constant on temperature can be described by exponential law obtained by Swedish chemist Svante Arrhenius empirically:

$$k(T) = Ae^{-\frac{E_a}{RT}} , \qquad (2)$$

where *k(T)* refers to the reaction rate constant with respect to temperature *T*, E_a is activation energy and *A* denotes frequency factor. It shows the probability that the molecules of reactants will have the correct orientation in order for a chemical reaction to occur.

Arrhenius equation in differential form is expressed as follows:

$$\frac{d \ln k(T)}{dT} = \frac{E_a}{RT^2} \qquad (3)$$

and in logarithmic form:

$$\ln k(T) = \ln A - \frac{E_a}{RT} . \qquad (4)$$

With at least two values of reaction rate constants with respect to different temperatures using (4) a system of equations is:

$$\begin{cases} \ln k(T_2) = \ln A - \dfrac{E_a}{RT_2}, \\ \ln k(T_1) = \ln A - \dfrac{E_a}{RT_1}, \end{cases} \qquad (5)$$

and then:

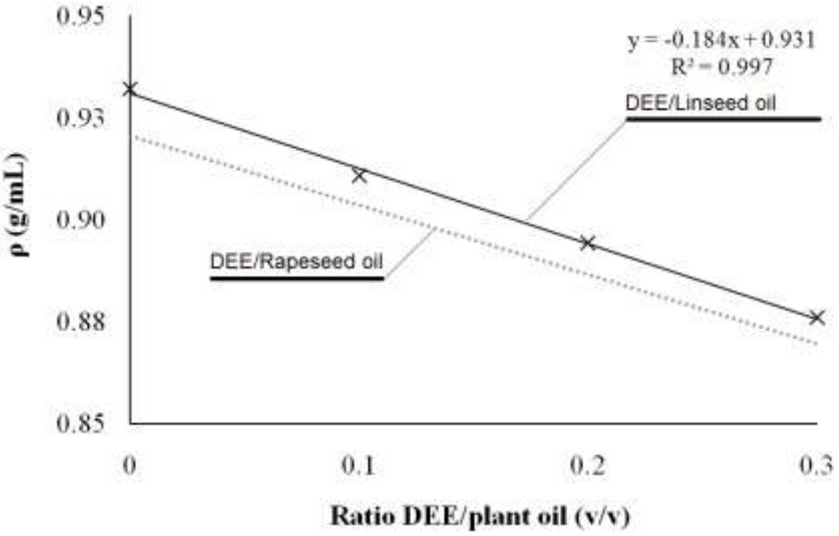
$$\ln \frac{k(T_2)}{k(T_1)} = \frac{E_a}{R}\left(\frac{1}{T_1} - \frac{1}{T_2}\right). \qquad (6)$$

Activation energy values can be calculated according to the formula:

$$E_a = R \cdot \frac{\ln \dfrac{k(T_2)}{k(T_1)}}{\left(\dfrac{1}{T_1} - \dfrac{1}{T_2}\right)} . \qquad (7)$$

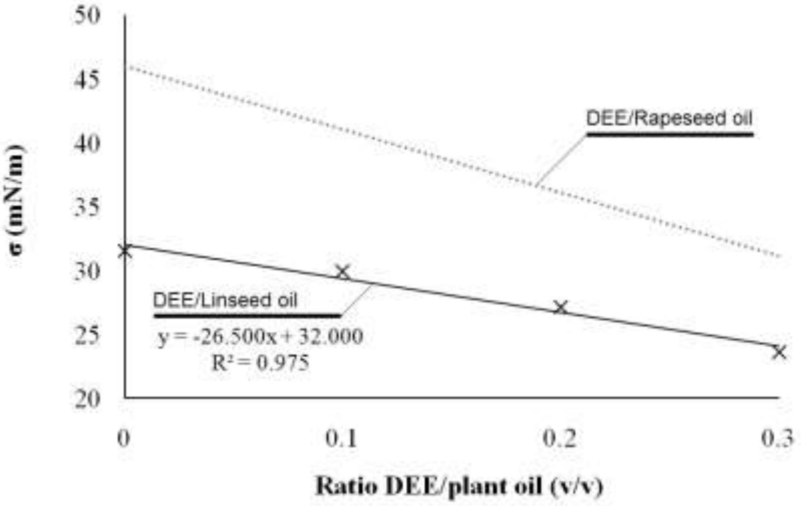
The resulting dependencies on the kinematic viscosity of fuel samples are shown in Fig. 1.

Density, as also viscosity, is temperature dependent parameter and there exist also dependence between viscosity and density, which is mainly used in oil industry. Like viscosity, also density has a great impact on the atomization process. Therefore, optimal values of those parameters must be reached for correct engine operation during all testing regimes. Results shown in Fig. 2 clearly indicate density dependence from DEE addition by a straight line with significant statistics outcomes in all cases with a confidence level of 95%. It could be observed that oil type does not leave a serious impact on density of all blends, therefore, LO can be used instead of RO to ensure optimal atomization process in diesel engine.



LO – continuous line; RO – dashed line

Figure 2 – Density (ρ) of DEE/LO and DEE/RO blend tested at 15 °C

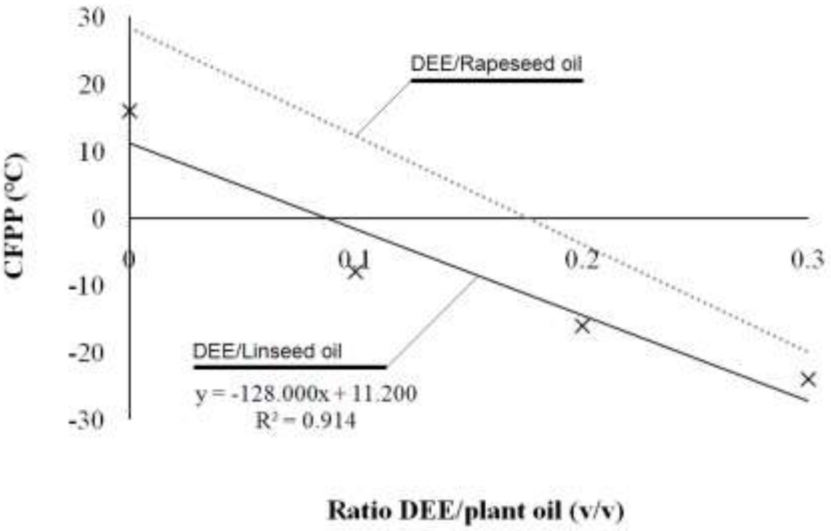


LO – continuous line; RO – dashed line

Figure 3 – Surface tension (σ) of DEE/LO and DEE/RO blend

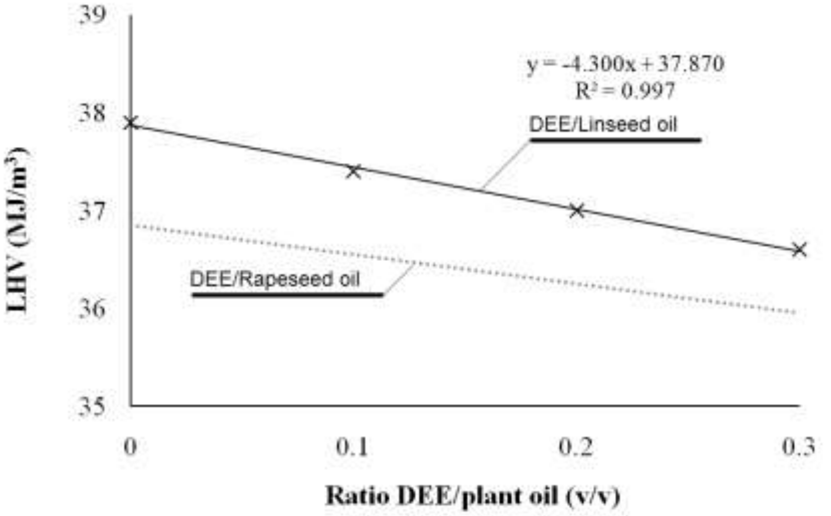
Another parameter, which could make atomization process difficult is surface tension. Although European standards do not define values of surface tension not in case of diesel fuel, neither to rapeseed oil, variation of this parameter leaves an important impact on injection process. High values of surface tension and viscosity ensure worse fuel atomization in the form of larger diameter spray droplets. It could be observed also in this research in case of RO, instead of LO, which ensures significantly lower values of surface tension without blending (Fig. 3). Like both previous parameters, also surface tension is temperature dependent, but it could be impacted also by pressure or even composition, like it is in case of blends. In this research could be observed linear decrease of surface tension based on DEE addition level to LO.

CFPP was another parameter, which was analysed in case of LO and DEE blends. As it is one of parameters, which is direct indicator of engine low temperature operability additional attention has to be turned by engine operation in problematic climate conditions. As it is seen in Fig. 4, LO indicates dependence from DEE by a straight line with tendency on reduction based on DEE addition. Even 10% addition to LO reduces CFPP to −8 °C, while RO can ensure such usage temperature only with 40% of DEE addition. In overall, addition of DEE improves low temperature properties for both oil types.



LO – continuous line; RO – dashed line

Figure 4 – CFPP (°C) of DEE/LO and DEE/RO blend



LO – continuous line; RO – dashed line

Figure 5 – Lower heating value (LHV) of DEE/LO and DEE/RO blend

Dependence of CFPP with respect to ratio of DEE/LO blend showed in Fig. 4 is expressed by linear regression $y = -128x + 11.2$ with R^2=0.914. However, to describe relationship between CFPP and DEE/LO ratio is better to use below showed, an irrational regression model $y = 15.766 - 72.493\sqrt{x}$, which better correlates with experimental data due R^2=0.997.

Results for LHV are presented in Fig. 5. Here it is possible to observe impact of fuel composition on changes of LHV. Like all other vegetable oils, also LO have reduced LHV compared with DF, but it also has increase of oxygen content by 10% while diesel almost do not have oxygen. Combination of LO with DEE allows to increase total value of oxygen in mixture resulting with decrease of LHV proportionally of added DEE. Results of LHV dependence from DEE addition is showed by a linear model (Fig. 5). Impact of DEE addition to RO on variation of LHV have similar tendency like in case of DEE/LO blends. Main difference is only a shift between values of LHV for LO and RO.

According to experimental data, density *(ρ)* of DEE/LO and DEE/RO blend tested at 15 °C, surface tension *(σ)* of DEE/LO and DEE/RO blend, and lower heating value (LHV) of DEE/LO and DEE/RO blend have a direct proportional, linear relationship presented in Figures 2, 3 and 5 respectively. In these cases, processing of experiment data was done using the least squares method. Since any measurement results contain random errors values y_i will be considered realizations of random variables.

For surface tension (σ) of DEE/LO and DEE/RO blend and lower heating value (LHV) of DEE/LO and DEE/RO blend similar calculations of confidence intervals of linear regression parameters were performed, which confirmed significance of the linear mathematical model.

In this work the selected parameters of combustion process were also investigated. Research confirmed that ignition delay (ID) of LO is bigger (0.8 –2 °CA) than in case for DF, but it was observed directly at lower speeds, while in other conditions the gap slightly narrow down (see Fig. 6). As it was mentioned previously, there is possible to observe impact of polyunsaturated fatty acids in LO allowing to reach such difference between LO and DF. Here it is seen an impact of physical delay influenced by fuel properties and composition. Addition of DEE reduces value of LO ignition delay together with viscosity resulting in wider spray pattern. As it can be seen from Fig. 6, ID of DF at larger engine speeds (2000 rpm) is similar to DEE/LO blends. Here it is possible to observe impact of cylinder temperature and pressure on chemical part of delay period, which is more pronounced at higher speeds.

Conclusions

Based on the literature review it can be stated that LO possesses a wide range of properties, which could make it a considerable alternative to other vegetable oils. Results of this study showed that the viscosity of LO is significantly lower in comparison with RO allowing to reduced amount of DEE added to plant oil. For example, LO containing 30% DEE has the same viscosity as RO containing 40% of DEE. It should be pointed out that both these mixtures allow to reach a viscosity value comparable to DF. The results confirmed that the kinematic viscosity of tested blends is well expressed by exponential regression based on the Frenkel-Andrade equation.

Research showed that DEE reduces the density of plant oils. However, values comparable to DF are possible only for mixtures containing more than 50% of DEE. Based on empirical data it was confirmed that necessary relationships are very well described by linear regression.

The linear model also describes well the impact of the DEE/LO ratio on the surface tension value. It was observed that DEE added to LO or RO reduces the surface tension of the blend. In the case of DEE/LO blend it should promote better atomization of the fuel injected into the combustion chamber. It is attributed to the lower surface tension of DEE/LO blend compared with DEE/RO.

At the same time, the addition of DEE to LO significantly reduces CFPP allowing to use of such mixtures in the winter season without the engine fuel preheating system. In this aspect, the DEE/LO blends are much more recommended compared with DEE/RO mixtures. CFPP of DEE/LO blends is also well described by a linear model. However, an irrational regression shown in this work better correlates with empirical data.

Research has confirmed that the addition of DEE slightly reduces the LHV of tested plant oils. In this case relationship between LHV and the DEE/LO ratio is very well described by the linear mathematical model. It should be pointed out that the LHV of the tested DEE/LO or DEE/RO blend is close to 36-38 MJ/m3, i.e. about 12% less compared with DF. For this reason, the engine performance should be adequately reduced. It means that the top power and torque of an unmodified engine fuelled with an alternative fuel blend will be lower compared with DF.

Based on the engine research, it was found that the ignition delay of LO is significantly increased compared with DF. This was attributed to a considerably lower CN of plant oil. However, it was confirmed that DEE is an effective cetane improver and it allows for to reduction of ignition delay of LO. Simultaneously, smoke emission is also adequately reduced due to the atomization quality of a less viscous blend is better, and the consequence is lower smoke emission and slightly higher engine overall efficiency.

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