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Environmentally Friendly Diesel Fuel Obtained from Vegetable Raw Materials and Hydrocarbon Crude

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Abstract: Currently, the global issue for countries is the search for raw materials and the production of bioenergy within their country; bioenergy also includes biodiesel fuels. One of the most promising biodiesel fuels is the green diesel fuel produced by the hydrogenation of vegetable oils. Three methods have been proposed to obtain high-quality biodiesel and environmentally friendly diesel fuel: compounding green diesel with hydro-treated diesel fuel, compositions of the improved fuel «green diesel» with bio-additives, and two-component mixtures of environmentally friendly diesel fuel with bio-additives. Using these methods, it is possible to produce fuel for diesel engines with improved lubricating properties, the wear scar diameter is reduced to 232 microns, according to EN 590: 2009, this value standard is up to 460 microns. The optimal quantitative composition of three-component environmentally friendly diesel fuel with improved lubricity was established. The dependence of the change in the lubricating properties of environmentally friendly diesel fuel on the quantitative and qualitative composition are established. A mathematical equation describing the dependence of the change in the corrected wear spot on the amount of anti-wear additive in the green diesel fuel is derived. Three-component compositions of environmentally friendly diesel fuel make it possible to obtain fuel that meets the requirements of the EN 590: 2009 standard and to expand the resources for obtaining fuel, as well as to improve the environmental and operational characteristics of the fuel.

Keywords: biodiesel; green diesel; fuel; bio-additive; lubricating properties; diesel

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Citation: Eremeeva, A.M.; Kondrasheva, N.K.; Khasanov, A.F.; Oleynik, I.L. Environmentally Friendly Diesel Fuel Obtained from Vegetable Raw Materials and Hydrocarbon Crude. *Energies* **2023**, *16*, 2121. https://doi.org/10.3390/en16052121

Academic Editor: Diego Luna

Received: 9 February 2023
Revised: 17 February 2023
Accepted: 21 February 2023
Published: 22 February 2023

1. Introduction

All over the world, one of the most urgent issues is air pollution and the reduction in the concentration of harmful emissions in the atmosphere to the minimum values. Carbon monoxide, nitrogen oxides, and hydrocarbon vapors, as part of exhaust gases, have the greatest impact on human health and nature [1–3]. Carbon monoxide enters the human body and builds up in the bloodstream, where gas molecules bind with hemoglobin molecules. This bond is stronger than the bond of oxygen molecules with hemoglobin molecules, therefore, the more carbon monoxide that is in the air of the working area, the less oxygen that can be retained in the blood of a person, as a result of which, oxygen deficiency can occur [4]. Nitrogen oxides are poisonous, irritate mucous membranes, and can cause pulmonary edema [5].

Not forgetting about the issue of greenhouse gases, programs are being developed around the world to decarbonize and reduce CO_2 gases to a minimum [6,7].

Another popular issue for many countries is the lack of oil resources for fuel production. Due to the lack of oil reserves and the low level of its processing (85%), European countries [8,9] need to create technologies for the production of alternative fuels [10–12]. Since most of the transport in these countries is equipped with diesel engines, the type of biofuel that can be used in them is the most in demand [13]. Biodiesel does not affect plants and animals when it contacts water. In addition, it undergoes almost complete biodegradation: in soil or in water, it allows microorganisms to process 99% of biodiesel in 28 days [14–16], which means the mitigation of the pollution of water-ways [17]. This fuel is 75% cleaner than petroleum diesel fuel [18,19].

The amount of biodiesel has been on the rise in recent years. Over the past decade, biodiesel production has increased approximately two times (Figure 1) [20].

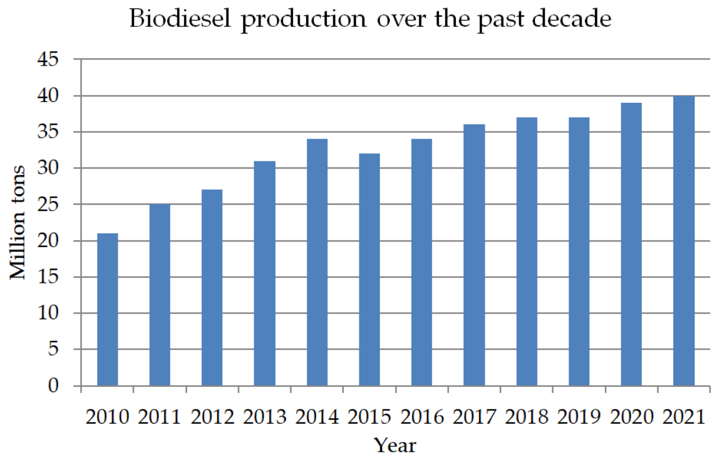

Figure 1. Biodiesel production over the past decade.

Recently, diesel vehicles gained popularity in Russia and Europe [21] due to the obvious advantages of renewable biodiesel, such as improved environmental properties (reduced emission of particulate matter and volatile organic compounds during combustion) [22,23], improved lubricity, and high cetane number.

2. The Overview of the Research Subject

In November 2022, the United Nations announced that the world's population had crossed the threshold of 8 billion people [24]. Constantly growing market needs are forcing manufacturers to increase production volumes, which leads to an increase in cargo transportations and, consequentially, to a higher level of consumption of petroleum products [25]. However, the reserves of easily recoverable oil are almost exhausted. The extraction of hard-to-recover oil (paraffin-based oil) is complicated both technologically and economically [26,27]. That is the reason why the search for alternative fuels has been a significant point of interest for a long time. For example, in [28,29], the authors considered the general principles for obtaining biodiesel from rapeseed raw materials. In [30], the authors analyzed the synthetic fuel market for the period 2020–2023 inclusive. The book [31] analyzes the technological process of obtaining biodiesel fuels. Their comparative characteristics and assessments of prospects are given. It is important to note that the technological process of obtaining biodiesel fuel is technologically very complicated. For example, the rapeseed drying must take place in a very limited temperature range, since over-drying or transportation errors can lead to a decrease in the percentage of oil in the grains, which can lead to a significant deterioration in the volume of fuel received. For such an adjustment, as a rule, numerical methods of distributed control systems [32,33] or analytical methods [34,35] are used. Using the methods above [36], it is possible to implement a complete technological chain for the production of biodiesel fuels [37,38]. A fairly small number of works are devoted to improving the quality of biodiesel fuels. A great number of publications are aimed at the one component of improving the fuel. However, they do not see the full picture. Many scientists [39,40] tend to believe that this has an economic basis: the unprofitability of fuel on the market is the main reason for the lack of the necessary improvement. Others [41,42] tend to consider the low level of the chemical industry in developing countries as the main problem for the lack of demand. The countries leading in the production of biodiesel fuels are shown in Figure 2 [14,17,20].

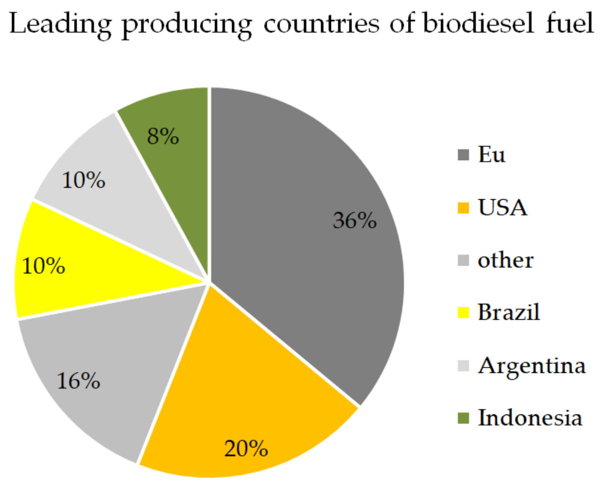

Figure 2. Leading producing countries of biodiesel fuel.

The lubricity of biodiesel fuels is discussed in [43–45]. It is important to note that lubricity cannot be obtained analytically. It can only be obtained experimentally. Therefore, to obtain it, numerous experiments are carried out in these works. However, neither these or other analyzed works provide a search for patterns of lubricity. Within the framework of this study, the task of assessing biodiesels lubricity and finding patterns has been considered.

3. Materials and Methods

There are several methods for obtaining environmentally friendly diesel fuel components, such as deep hydrotreating of the straight-run diesel fraction, transesterification of vegetable oils with alcohols, esterification of fatty acids with alcohols, hydrotreating of vegetable oils over a catalyst, catalytic cracking of vegetable oils, and applying of vegetable oils in pure form. It is most rational to use only a few of them to obtain high-quality products, specifically transesterification of vegetable oils with alcohols, and hydrotreating of vegetable oils over a catalyst. In the first case, a mixture of carboxylic acid esters is obtained, the advantages of which are an increased cetane number, an increased flash point in a closed crucible, and improved lubricity. The serious disadvantages of this type of fuel include increased oxidative stability of the fuel due to the fact that the composition contains oxygen-containing compounds. In the second case, a mixture of paraffins of normal and iso-structure is obtained. The advantage of this composition is the high cetane number and similitude in physico-chemical properties to diesel fuel, however, the process itself is expensive, and the low-temperature and lubricating properties do not meet the requirements of standards and require improvements. On the economic side, it is most profitable to use vegetable oil in its pure form, there is no need to attract additional capital and manufacturing investments for raw materials, equipment, etc., but due to high viscosity and the formation of a large amount of soot and solid particles, incomplete combustion of oils occurs, which quickly clogs and disables the engine.

Thus, in the later part of the study, biodiesel fuel obtained by hydrotreating and also transesterification of vegetable oil was investigated.

Experimental Section

Green diesel was obtained using the process of hydro-processing vegetable oils [46,47]. The formation of a mixture of C_{15}–C_{18} liquid hydrocarbons boiling off within the diesel fractions is realized due to the occurrence of decarboxylation, hydrodecarbonylation, and "reduction" reactions, and is accompanied by the release of carbon oxides (CO_2, CO) and water [48,49]. Under hydrocracking conditions, other light alkanes (methane, ethane, butanes) are formed as a result of the breaking of C–C bonds along with propane. In this case, liquid hydrocarbon products are enriched with C_5+ alkanes with boiling points within the boiling range of gasoline and jet fuels.

Using a gas chromatography–mass spectrometer GCMS-QP2010 SE Shimadzu [50], a quantitative and qualitative composition of biofuels was obtained. The green diesel sample contains only 88 compounds.

Biofuel consists mainly of n-alkanes and iso-alkanes (Figure 3). This composition explains the higher pour point of the fuel compared to HTDF. The difference in pour points is explained by the fact that the solidification temperature of diesel fuel depends on the content of paraffin hydrocarbons, which are mainly contained in green diesel fuel and have a high melting point. The HTDF contains a mixture of various hydrocarbons and the proportion of paraffin is significantly less than that of green diesel fuel. Also, this composition has poor lubricity, as there are no heteroatomic compounds in the composition that can reduce friction in engine parts [51]. However, this composition causes a high cetane number of fuel and environmental safety—the minimum amount of harmful emissions from fuel combustion in engines. The cetane number is one of the most important operational characteristics of diesel fuel, which is determined primarily by the composition and shows the time interval from fuel injection into the cylinder to the combustion initiation. The higher the cetane number, the lower the delay and the more calmly and smoothly the fuel mixture burns, while a higher cetane number also ensures complete combustion of the fuel mixture, rapid ignition, and the formation of a small amount of soot. The largest cetane number is possessed by paraffin hydrocarbons, which are contained in large quantities in green diesel fuel.

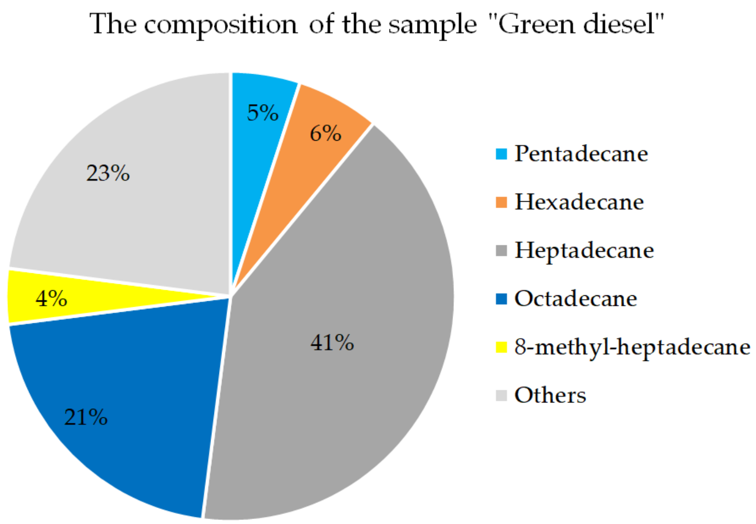

Figure 3. Qualitative and quantitative composition of green diesel.

The basic physicochemical and operational properties of biodiesel were also identified, as presented in Table 1.

Based on the physicochemical characteristics of the green diesel fuel (Table 2), it can be concluded that in order to use this type of fuel in a diesel engine, it is necessary to improve, primarily, the lubricating properties of the fuel.

One of the main tasks of the research was to improve the lubricating properties of the green diesel fuel. There are several ways to improve the lubricity of diesel fuels [52], the main of which are:

- Compounding with components with better lubricity;
- The use of bio-additives with improved lubricating properties [53,54].

A second way to improve the lubricating properties of the fuel is the introduction of anti-wear bio-additives in the fuel.

Table 1. Physicochemical characteristics of HTDF and the green diesel sample.

№	Name of Indicator	Green Diesel	HTDF
1	Density at 15 °C, kg/m^3	777.1	854.8
2	Sulfur content, mg/kg	36	8
3	Flash point in a closed crucible, °C	90	80
4	Lubricity: adjusted diameter of the wear spot at 60 °C, microns	595	443
5	Kinematic viscosity at 40 °C, mm^2/s	3.48	2.79
6	Fractional composition: - at a temperature of 250 °C, % (by volume) - at a temperature of 350 °C, % (by volume), - 95% (by volume) is distilled at a temperature, °C	 4 94 298	 123 213 330
7	Cetane number calculation method	70	51
8	Pour point, °C	18	−17
9	The content of methyl esters of fatty acids, % (by volume)	-	-
10	Ash content, % by weight	0.008	-
11	Water content, mg/kg	-	-
12	Total contamination, mg/kg	-	-
13	Copper strip corrosion, rating	1	1
14	Oxidation stability, g/m^3	11	16

Table 2. Anti-wear additives abroad.

	DF	HTDF	Anti-Wear Additives					Our Sample
			Kerokorr LA 99C	Dodilube 4940	PC 32	Hitec 4140A	R690	
Manufacturer			**BASF**	**Clariant**	**Total**	**Afton Chemical Corporation**	**Infineum**	
Sulfur content, ppm	220	5	12	9	15	7	2	10
CWSD, microns	300	650	400	307	330	382	201	113
Compound			Talic Acids				Glycerol esters	

As the main components designed to improve the lubricity of the fuel, carboxylic acids and their derivatives (esters and amides) are most often used. The composition depends on the nature of the processed raw materials. Fatty acids are a mixture of normal saturated and predominantly unsaturated acids with the C_{16}–C_{24} carbon chain (oleic, linolenic, linoleic, etc.).

To improve the lubricating properties, a bio-additive based on sunflower oil and dihydric alcohol was developed [55,56]. In the process of obtaining additives, various vegetable oils (both edible and non-edible) can be used as raw materials, because triglycerides mainly contain linoleic, linolenic, and oleic acids in various proportional ratios. The choice of the authors was due to the availability of raw materials and low cost. As the alcohol, monohydric alcohols, dihydric alcohols, and trihydric alcohols can also be used. Our final choice was due to the maximum degree of conversion of raw materials, as well as economic feasibility.

In the world, the main types of anti-wear additives are Kerokorr LA 99C and Dodilube 4940 [57–59]. As can be seen, the main components of these additives are tall oils, which significantly reduces the cost of technology and the cost of the finished product and glycerol esters [21,60–64]. However, acids, even in small amounts, adversely affect the oxidizing ability of the fuel [65–67] (Table 2).

The main components of the additive we obtained are fatty acid esters contained in vegetable oil. Using a chromatography-mass spectrometer Agilent 5973 in a non-polar DB-Petro column with a length of 100 m, a quantitative and qualitative composition of the synthesized compounds was obtained (Table 3). The number of esters amounts to 77.78% of the mass.

Table 3. The composition of the ester part of bio-additive.

№	Ester Name	Content, %
1	Linoleic acid methyl ester	1.21
2	Oleic acid methyl ester	0.49
3	Linoleic acid ethyl ester	1.05
4	Linoleic acid 2,3-dihydroxypropyl ester	38.75
5	Elaidic acid hydroxyethyl ester	24.48
6	Glycerin ester of elaidic acid	6.53
7	Stearic acid hydroxyethyl ester	2.75
8	Linoleic acid 2,3-dihydroxypropyl ester	1.37
9	Tranexamic acid cetyl ether	0.73
10	Linoleic acid ethyl ester	0.42

According to Table 3, it is seen that the main acids that enter into the esterification reaction are oleic (or its isomer) and linoleic.

4. Results and Discussion

To create compositions of environmentally friendly diesel fuel with improved lubricating properties, green diesel fuel was compounded with other components.

4.1. Compounding Green Diesel with Hydrotreated Diesel Fuel

As mentioned earlier, one of the ways to reduce the diameter of a fuel wear spot is to compound a green diesel fuel with hydrotreated diesel fuel (HTDF) [55], which has better lubricating properties (595 microns versus 443 microns, respectively). The characteristics of the hydrotreated diesel fuel used are presented in Table 1.

The following compositions of blended environmentally friendly diesel fuel were compiled and their adjusted diameter of the wear spot was determined (Table 4).

Table 4. Two-component superior diesel formulations.

Component	Amount of Component, %										
Green diesel, %	100	90	80	70	60	50	40	30	20	10	0
HTDF, %	0	10	20	30	40	50	60	70	80	90	100
CWSD *, microns	595	590	583	572	568	563	550	492	420	370	443
CWSD, EN 590: 2009, microns	460										

* CWSD—corrected wear spot diameter, microns.

Based on the results obtained (Table 2), we can conclude that not a single mixture of HTDF with the green diesel sample as a fuel component passes according to the requirements of EN 590: 2009 "Diesel fuel EURO. Technical conditions", therefore, the introduction of anti-wear bio-additives is necessary for the use of this type of fuel in diesel engines. Most authors in their articles [28,38–40] suggest using green diesel fuel as the main fuel or component. Most often, hydrotreated diesel fuel, which is the second component in a mixture with green diesel fuel, has a lubricity index above the maximum value specified in the requirements of the standards, therefore, the use of anti-wear additives is necessary for such mixtures. In our case, the lubricity index of hydro-treated diesel fuel satisfies the requirements of the standard and is slightly below the specified maximum value, therefore, this method can be used to improve lubricity, however, as the results show, it is insufficient to achieve the desired result.

The dependence of lubricity on the quantitative content of hydro-treated diesel fuel in a mixture with green diesel fuel is described by a power function of the fifth degree, presented below (1). The value of the reliability of the result is 99.93%.

$$y = 1 \cdot 10^{-6} \cdot x^5 - 0.0003 \cdot x^4 + 0.026 \cdot x^3 - 0.9223 \cdot x^2 + 13.25 \cdot x + 526.58 \quad (1)$$

4.2. Compositions of the Improved Fuel Containing «Green Diesel» and Bio-Additives

Bio-additives to the green diesel fuel were introduced in an amount of 5 to 50%, and the results were compared with the standard for diesel fuel EN 590: 2009 (Table 5).

Table 5. Dependence of lubricity on the content of bio-additives in the green diesel fuel.

Component	Content, %											
Green diesel	100	95	90	80	70	60	50	40	30	20	10	0
Bio-additive	0	5	10	20	30	40	50	60	70	80	90	100
CWSD, microns	595	232	327	247	294	335	238	298	273	267	324	536
CWSD, EN 590: 2009, microns	460											

The best results are shown in formulations containing 5 and 50% bio-additives (232 and 238 microns, respectively). Nevertheless, it is most effective to use a sample with a dietary supplement content of 5%, since the SDPI is almost at the same level, and the amount of dietary supplement in the mixture is an order of magnitude lower. In addition, for the normal operation of a diesel engine without significant changes in its design, it is recommended to use a bio-additive in fuel in a small amount not exceeding 5%. This dependence cannot be described by a power function with a sufficient degree of reliability of the results. Graphically, the dependence of the lubricity of the fuel on the amount of bio-additives in the green diesel fuel is shown in Figure 4. Most authors believe that this dependence is either hyperbolic in nature with an increase in the amount of bio-additives, the corrected diameter of the wear scar decreases [68–70], or an extreme nature with one minimum [71]. In most cases, it does not matter what raw materials are used to obtain bio-additives [72,73].

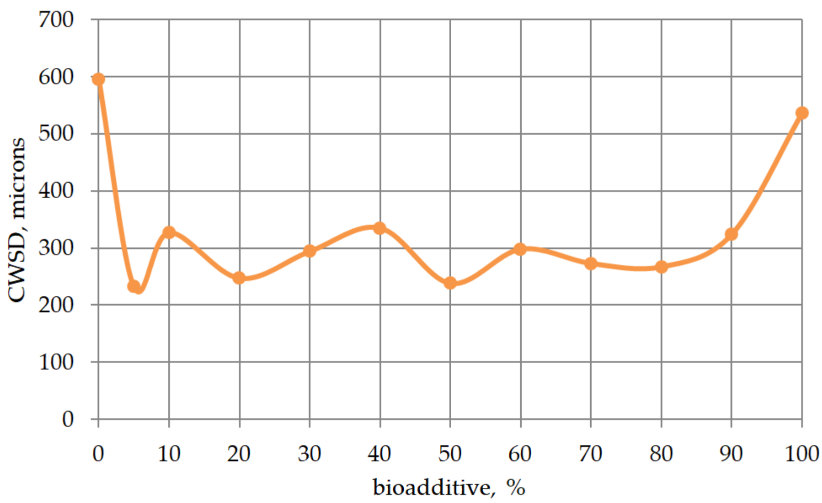

Figure 4. Dependence of lubricity on the content of bio-additives in the green diesel fuel.

Thus, the graphof the dependence of the lubricating ability of environmentally friendly diesel fuel "Green diesel" on the content of bio-additives in it is extreme (Figure 4), with a minimum in the range from 5 to 50% content of bio-additives, since with increasing concentration of biofuel up to 5% in a mixture with green diesel, adsorption of heteroatoms (oxygen molecules with unpaired electrons) takes place on the metal surface of the rubbing pair of a metal ball—a plate immersed in the test fuel, and is strengthened by the intermolecular interaction at the metal–liquid phase boundary. With an increase in the biofuel concentration in the mixture over 50%, the metal–liquid phase interface is supersaturated with heteroatoms, there is a coagulation (coarsening) and desorption of larger molecules into the solution volume, and, consequently, a decrease in the intensity of intermolecular interaction of the fuel with the metal and an increase in the CWSD.

4.3. Two-Component Mixtures of Environmentally Friendly Diesel Fuel with Bio-additives

After that, to improve other fuel characteristics (pour point, kinematic viscosity, etc.) at the same time, it was decided to compile environmentally friendly diesel fuel (EFDF) mixtures consisting of two basic components (green diesel and HTDF) and synthesized bio-additives.

To present the results of experimental studies, several graphical dependences of the lubricity of various fuel compounds with bio-additives were constructed, in which the amount of one or two components remained unchanged.

Compositions of environmentally friendly diesel fuel with bio-additives were developed, containing 5% and 10% bio-additive, and the amount of green diesel sample varied from 25 to 45%, while the remaining fuel was HTDF.

As can be seen from Table 6, all the mixtures obtained satisfy the requirements of the standard EN 590: 2009 (no more than 460 microns) in terms of lubricity, but the best result for a two-component composition with bio-additive in the amount of 10% is shown by the fuel composition consisting of 30% of the green diesel and 60% HTDF, as well as clean starting components of the base fuel with 5% bio-additive and HTDF (95%)—113 microns.

Table 6. Dependence of lubricity on changes in the component composition of mixed fuel with the amount of bio-additive 5% and 10%.

Green Diesel, %	HTDF, %	CWSD, Microns	CWSD, EN 590: 2009, Microns
Amount of bio-additive 5%			
95	0	232	460
45	50	326	
35	60	267	
25	70	279	
0	95	113	
Amount of bio-additive 10%			
90	0	327	460
40	50	270	
30	60	233	
0	90	133	

Thus, with an increase in the amount of the green diesel component (that is, with a decrease in the amount of HTDF) in the composition of EFDF (with a bio-additive content of 5 wt%), there is a general tendency for the lubricity of mixed fuels (i.e., an increase in CWSD) to deteriorate with a small extremum in the concentration range of 50–70% HTDF in mixed fuel.

5. Conclusions

In a rapidly growing economy, the issue of obtaining alternative, environmentally friendly fuels is acute. This paper considers the solution of an important scientific and technical problem—improving the quality of biodiesel fuel. Lubricity was chosen as the object of a study.

From the obtained data, it can be concluded that the green diesel sample cannot be used without anti-wear bio-additives, both in pure form and as a component of environmentally friendly fuel mixed with hydrotreated diesel fuel, because the lubricity of the mixtures obtained does not meet the requirements of EN 590: 2009, since with an increase in the amount of green diesel fuel the lubricity of the fuel deteriorates, and the diameter of the fuel wear spot increases.

With introduction of anti-wear additives and an increase in their concentration in the green diesel fuel and in mixed environmentally friendly diesel fuel (hydro-treated diesel fuel with green diesel fuel), the lubricity of the samples improves (the wear spot diameter decreases from 595 to 233 microns)

The best composition of green diesel fuel and bio-additive contains 95 and 5%, respectively. The diameter of the wear spot in this case is 232 microns. The best composition of mixed environmentally friendly diesel fuel with bio-additive contains 60% HTDF, 30% green diesel fuel, and 10% bio-additive, and the diameter of the wear spot is 233 microns.

The developed technology for the production of environmentally friendly diesel fuel from vegetable and petroleum raw materials with improved lubricating properties will expand the scope of fuel resources for production of motor fuels for diesel engines, as well as give an opportunity to produce biodiesel and environmentally friendly diesel fuels that meet international standards. Production of high-quality environmentally friendly diesel fuel will reduce the dependence between countries on the level and quantity of oil production.

The article presented possible solutions to the problem of the extension of diesel engines lifetime.

Author Contributions: Conceptualization, A.M.E.; methodology, A.M.E. and I.L.O.; investigation, A.M.E.; writing—original draft preparation, review and editing, A.M.E., N.K.K. and A.F.K. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Data Availability Statement: All data are available in the main text.

Conflicts of Interest: The authors declare no conflict of interest.

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