POLLUTION

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METHODS FOR CHANGING THE COMPOSITION AND PROPERTIES OF BIODIESEL

Research article

Abstract

The widespread use of engines using petroleum products has led to increased pollution of air, soil and water. In modern conditions, biodiesel is a good alternative to petroleum fuels, which has several advantages. Biodiesel is biodegradable, nontoxic and environmental friendly. The use of biodiesel makes it possible to improve the ecological situation, reduce the environmental anthropogenic impact and decrease the emissions of carbon oxides, polycyclic aromatic hydrocarbons and sulfur oxides into the environment. Use of biodiesel facilitates storage and transportation of fuel and liquidation of accidental or emergency spills. However, biodiesel physicochemical properties do not quite meet the requirements for auxiliary diesel fuels. To change the properties of biodiesel, it is necessary to modify its composition. We have established the dependence of the properties of biodiesel on its composition, as well as the dependence of certain physicochemical parameters on temperature. Methods for changing the composition, and, consequently, the properties of biodiesel, are proposed: mixing with individual low molecular weight esters, gene modification of oil-containing plants, both higher and microalgae.

Keywords: biodiesel, environment, physico-chemical properties, biodiesel composition, genetic engineering.

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СПОСОБЫ ИЗМЕНЕНИЯ СОСТАВА И СВОЙСТВ БИОДИЗЕЛЬНОГО ТОПЛИВА

Научная статья

Аннотация

Широкое применение двигателей, использующих нефтепродукты, привело к повышенному загрязнению воздуха, почвы и воды. В современных условиях биодизельное топливо является хорошей альтернативой нефтяному топливу, имея ряд преимуществ. Биодизельное топливо является биоразлагаемым, нетоксичным и экологически чистым. Использование биодизельного топлива позволяет улучшить экологическую ситуацию, уменьшить антропогенное воздействие на окружающую среду и снизить выбросы оксидов углерода, полициклических ароматических углеводородов и оксидов серы в окружающую среду. Использование биодизеля облегчает хранение и транспортировку топлива и ликвидацию аварийных или аварийных разливов. Однако физико-химические свойства биодизельного топлива не вполне соответствуют требованиям к современному дизельному топливу. Чтобы изменить свойства биодизеля, необходимо изменить его состав. Установлена зависимость свойств биодизельного топлива от его состава, а также зависимость некоторых физико-химических параметров от температуры. Предложены методы изменения состава и, следовательно, свойств биодизеля: смешивание с индивидуальными низкомолекулярными эфирами, генная модификация маслосодержащих растений, как высших, так и микроводорослей.

Ключевые слова: биодизельное топливо, окружающая среда, физико-химические свойства, состав биодизеля, генная инженерия.

1. Introduction

The active development of industry, agriculture and the transport sector make the problem of using fuel resources of current interest. For diesel engines of tractors and other agricultural machines, trucks and cars diesel oil is mainly consumed. The major disadvantage of using petroleum based fuel is atmospheric pollution. Petroleum originating fuels are regarded as environment-hostile due to their influence on carbon dioxide accumulation in the atmosphere leading to the development of the greenhouse effect. Moreover, petroleum resources are constantly decreasing, and their renewal is very slow. Continued growth in world oil prices and a growing shortage of oil products lead to the need for alternative motor fuels. It is critical to focus on renewable resources and development of new technologies [1-8].

Currently, biodiesel is becoming popular as an alternative environment friendly fuel. The consumption of biodiesel has experienced an impressive growth during the last years. Biodiesels are mono-alkyl esters of vegetable oils or animal fats that are produced by trans-esterification reactions with alcohols. Biodiesels can be the fuel of the future as economical, eco-friendly, and renewable energy source.

The advantages of biofuel over the conventional diesel fuel:

– raw materials for the synthesis of biodiesel are vegetable oils, the components of which are not hydrocarbons, but esters of glycerol and aliphatic carboxylic acids;

– in the molecules of esters there are oxygen atoms, which contributes to a more complete combustion of fuel, low carbon dioxide and carbon monoxide emissions;

– aromatic and sulfur-containing substances are practically absent in oils, therefore, emissions of polycyclic aromatic and unburned hydrocarbons and sulfur to the atmosphere are reduced;

– biodiesels have a higher flash point, high cetane numbers and good lubricating properties;

– biodiesel is biodegradable, nontoxic and suitable for sensitive environments. This reduces the environmental anthropogenic impact, facilitates storage and transportation of fuel and liquidation of accidental or emergency spills.

However, the wider use of biofuels based on vegetable oils is constrained by the differences of their physicochemical properties from those of petroleum diesel fuels, which can lead to deterioration in the operation of a diesel engine that uses biodiesel. Despite all the advantages, biodiesel is used at best as an additive to commercial oil. This is due to the fact that the esters that make up biodiesel have a relatively high molecular weight comparing to hydrocarbons from petroleum fuels, and accordingly a higher density, viscosity. These differences can degrade the performance of an engine using such fuel, and increase the varnish and deposit deposition. A higher molecular weight of the components leads to a deterioration in the low temperature properties of the fuel. The high viscosity of biodiesel requires modernization of the engine (for example, adding a device for heating the fuel before it enters the combustion chamber). The presence of unsaturated components determines higher iodine number, and causes a greater reactivity of biodiesel esters and reduces their storage stability. Therefore, much attention is paid to finding the best ratio of biodiesel and petroleum fuel to produce mixed fuel having such performance characteristics that would not require modernization of the engine [9-12].

However, in order to improve the physicochemical and operational characteristics, the modification of biodiesel composition seems the most promising and rational.

This scientific research has the purpose to analyze the correlation between the physicochemical and operational properties of biodiesel and its composition, and choose effective methods that allow changing and improving its characteristics.

2. Methods

The synthesis of individual esters of aliphatic acids and methyl alcohol was carried out according to the esterification reaction; sulfuric acid was used as a catalyst. The synthesis of biodiesel was carried out according to the transesterification reaction. Potassium hydroxide was used as a catalyst. The following vegetable oils were used as a raw material for synthesis: rapeseed, linseed, corn, sunflower, camelina, and radish oil. Physico-chemical properties of biodiesel were determined in accordance with GOST R 52368-2005. "Diesel fuel EURO. Technical conditions". The fatty acid composition of biodiesel was determined by Crystal 2000m gas chromatograph. Chromatek Analytic software was used to process the results of chromatographic analysis.

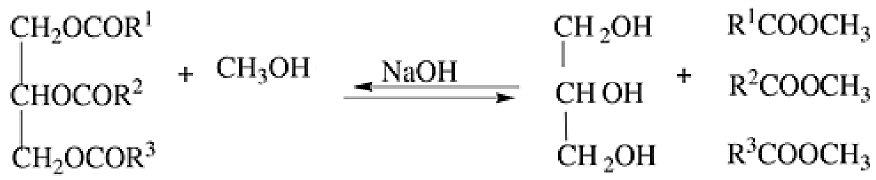
3. Discussion

The study of the structure of biodiesel components and the relationship between the structure and the physicochemical and operational properties of fuels is necessary for the understanding how to change the composition of biodiesel. The components of biodiesel are methyl fatty acid esters. These acids are components of triacylglycerols of the vegetable oils used for biofuel synthesis.

Biodiesel is usually produced from oils of those plants that are common for a particular region. Thus, sunflower oil is one of the most common in Russia, in European countries rapeseed oil is of more interest, soybean oil is widely used in the USA, coconut or palm oil or non-food jatropha oil are used in countries with a tropical climate (India, Indonesia, Philippines), castor oil – in Brazil. Presently, non-edible oils, such as rancid and used deep-frying oils, microalgae lipids, etc. becoming increasingly important [13], [14].

Various plants produce oils differ in amount of triacylglycerols and their fatty acid composition. Besides of that, the variety of the terrain and climatic conditions affects the fatty acid composition of the oil of the same plant. Therefore, the biodiesel fuel synthesized from them will differ in the composition and in physicochemical and operational properties.

In the course of our investigation, several biodiesel samples from various vegetable oils obtained from agricultural crops growing in the middle zone of the Russian Federation, were synthesized by transesterification with methyl alcohol, and analyzed. The scheme of transesterification reaction is presented below.



The compositions of synthesized biodiesels are presented in table 1.

Table 1 – The composition of biodiesel synthesized from various oils

Biodiesel composition	The content of methyl esters of higher fatty acids in biodiesel samples synthesized from oils of, %					
	radish	sunflower	corn	rapeseed	camelina	linseed
$C_{13}H_{27}COOCH_3$	10,64	4,56	–	0,33	0,29	7,56
$C_{14}H_{29}COOCH_3$	–	–	–	–	–	3,65
$C_{15}H_{31}COOCH_3$	11,85	8,61	8,31	3,99	5,65	6,46
$C_{15}H_{29}COOCH_3$	–	0,60	–	–	–	–
$C_{17}H_{35}COOCH_3$	3,53	4,77	6,17	–	–	2,25
$C_{17}H_{33}COOCH_3$	31,82	49,25	47,09	59,53	67,46	7,89
$C_{17}H_{31}COOCH_3$	24,59	18,90	37,43	20,29	2,39	23,32
$C_{17}H_{29}COOCH_3$	9,38	8,42	0,53	6,03	–	46,73
$C_{19}H_{39}COOCH_3$	–	3,43	0,15	4,32	17,9	2,14
$C_{19}H_{37}COOCH_3$	–	0,34	0,32	0,66	1,6	–
$C_{21}H_{43}COOCH_3$	–	1,73	–	0,25	0,59	–

Biodiesel produced from rapeseed and corn oils is characterized by high content of oleic and linoleic acid esters (around 80%), while linseed and radish-oil based biodiesel is rich in low molecular esters (myristic acid ester) and polyunsaturated esters (linoleic and linolenic acid esters).

Transesterification of oil and fats using short chain alcohols, results in monoesters having viscosity that is closer to petroleum based diesel fuel [15]. The properties of biodiesel samples from various vegetable oils are presented in table 2.

Table 2 – Physico-chemical parameters of biodiesel samples obtained from various oils

Parameter	Values for the samples received from oils of:					
	radish	sunflower	corn	rapeseed	camelina	linseed
Density15°C, kg/m^3	876	880	880	882	884	888
Kinematic viscosity 20 °C, mm^2/s	7,41	6,59	6,77	6,77	6,48	6,77
Kinematic viscosity 40 °C, mm^2/s	4,94	5,08	4,97	5,00	–	4,44
Cetane number	–	53	52	53	–	53
Pour point, °C	- 12	- 7	- 8	- 8	- 11	- 9
Cloud point, °C	+ 3	- 1	- 1	0	- 4	- 3
Flash point, (in a closed crucible), °C	121	140	152	161	158	153

Analysis of the data of tables 1 and 2 shows a correlation between the fatty acid composition and the physicochemical properties of biodiesel.

Not all biodiesel fuel parameters meet the requirements for commercial diesel fuel, a slow temperature properties, density, and viscosity. It is possible to improve biodiesel characteristics by using mixtures of biodiesel and petroleum fuels. Another way is addition of low molecular weight esters. Blending of different methyl esters can show significant improvement in biodiesel properties. It allows reduce biodiesel viscosity and density; make the fractional composition wider, bringing it closer in properties to oil fuel, and increase biodiesel stability. These low molecular esters are obtained by the esterification reaction from petroleum or renewable plant components.

With the introduction of low molecular weight esters, the low temperature properties of biodiesel are also improved, and the advantage in terms of lubricity and environmental properties is preserved [16].

Thus, biodiesel from sunflower, rapeseed and corn oils with high content of esters of linoleic and oleic acids have close values of density, viscosity, pour point and cloud point.

The highest content of linolenic acid esters (with three double bonds in the radical) of biodiesel from linseed and radish oils result in the lowest viscosity.

The temperature dependence of the density of biodiesel fuels synthesized from various oils is presented at Fig. 1.

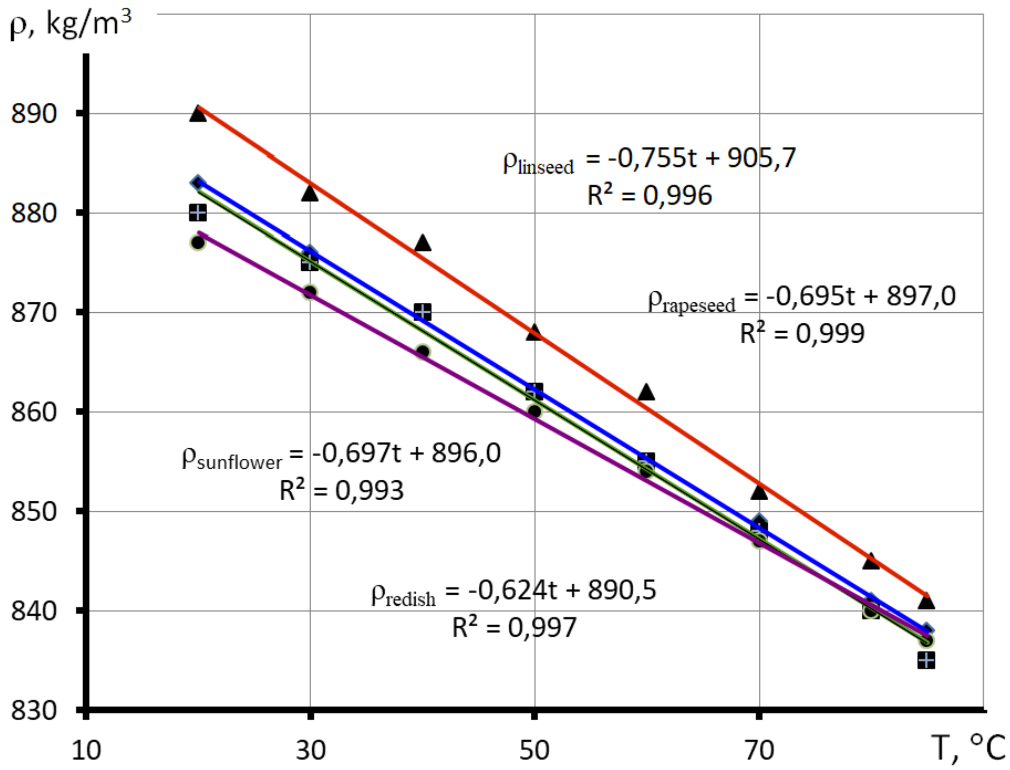


Figure 1 – Experimental dependence of the density of biofuels synthesized from various oils on temperature

It is known, that the dependence of density on temperature is linear for vegetable oils and petroleum diesel fuels. The same result was obtained for synthesized biodiesel fuels (Fig. 1). The difference in the densities of the biodiesel samples differ in their fatty acid composition is clearly seen, but the angle of inclination of the trend lines is almost the same.

The viscosity dependence of the density of biodiesel fuels synthesized from various oils is presented at Fig. 2.

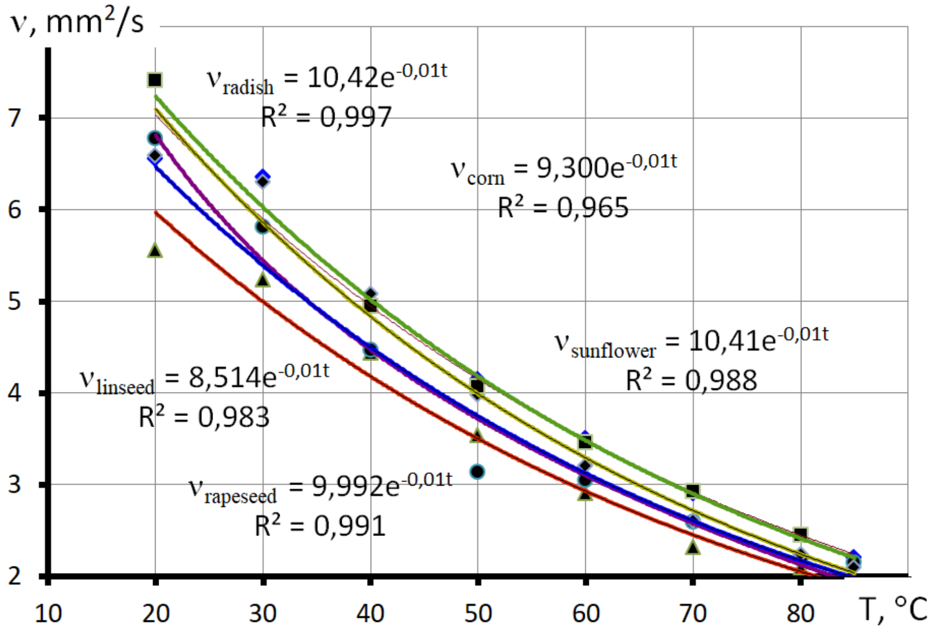


Figure 2 – Experimental dependence of the kinematic viscosity of biofuels synthesized from various oils on temperature

The viscosity of vegetable oils has an exponential dependence on temperature [17], while a power function is best suited for hydrocarbons from petroleum diesel fuels. The dependencies presented in Fig. 2 for synthesized biodiesel fuels are also best described by an exponential function. This difference is apparently due to the fact of different chemical nature of biodiesel and vegetable oils molecules. Vegetable oils are esters in nature, and oils contain hydrocarbons. This fact determines different intermolecular interactions and thus different types of viscosity dependence on temperature.

As biodiesel is a multi-component mixture, it was a subject of interest to analyze properties of individual components. Using esterification reaction of higher acids and methyl alcohol, we synthesized and analyzed several samples of methyl esters of individual fatty acids. Formulas of esters and their physicochemical parameters are presented in Table 3.

Table 3 – Physicochemical parameters of methyl esters of individual fatty acids

Substance Formula	Melting Point, °C	Boiling Point, °C	Viscosity, 40°C, mm^2/s	Density (15°C, kg/m^3)	Cetane Number [1]	Heat of Comb. (kg·kal/mol)
$C_7H_{15}COOCH_3$	-40	193	1,18	881	33,6	1313
$C_9H_{19}COOCH_3$	-12,8	228	1,71	876	47,2	1625
$C_{11}H_{23}COOCH_3$	5	266	2,41	873	61,4	1940
$C_{13}H_{27}COOCH_3$	18,5	286	3,23	–	66,2	2254
$C_{15}H_{31}COOCH_3$	30,5	416	4,36	–	74,5	2550
$C_{17}H_{35}COOCH_3$	39	442	4,74	–	86,9	2859
$C_{17}H_{33}COOCH_3$	-20	216	4,40	877	55	2828
$C_{17}H_{31}COOCH_3$	-35	207	3,56	888	42,2	–
$C_{17}H_{29}COOCH_3$	<-35	207	3,14	901	–	–

Increasing chain length results in higher melting point and higher cetane number. Increasing unsaturation results in lower melting point and lower cetane number. The presence of a double bond reduces the melting and boiling points of methyl esters of unsaturated acids in comparison with the corresponding derivatives of saturated acids. Therefore, an increase in the fraction of unsaturated fuel components improves its low temperature properties.

The use of fuels with a very high concentration of unsaturated components is undesirable since heating of such type biodiesel accelerates the polymerization reaction, which can lead to the formation of deposits or to a deterioration in the quality of the lubricating oil. Increasing unsaturation leads to decreased oxidative stability and makes biodiesel less resistant to oxidation during storage. This effect increases with the number of double bonds in the chain of fatty acids (from one in oleic acid to three in linolenic). Therefore, for example, linseed oil with high contents of linolenic acid is not the most suitable raw material for biodiesel synthesis. The presence of high contents of monounsaturated oleic or palmitoleic acids in biofuel is most preferred.

The introduction of low molecular weight to improve properties of biodiesel is promising, but this will require the creation of a separate technology for carrying out the esterification reaction between low molecular weight alcohols and acids, as well as expanding the raw material base of biodiesel production, including through refined products.

However, the composition of biodiesel can be changed in another way. The length of the carbon chain, the degree of unsaturation and branching can be changed both by plant breeding and genetic engineering approaches, and by chemical treatment of biodiesel to split some double bonds or form branched isomers. However, there is very little research in this direction.

The desaturation of stearic acid (C18:0) to oleic acid (C18:1) is catalyzed by stearoyl-acyl carrier protein desaturase (SAD).The biological switch of oleic acid to linoleic acid is facilitated by fatty acid desaturase 2 enzyme (FAD2). Further desaturation of oleic acid to linoleic acid (18:2) is catalyzed by FAD2 in the endoplasmic reticulum and FAD6 in the plastid, whereas linoleic acid desaturation to γ-linolenic acid (C18:3) is catalyzed by FAD3 in the endoplasmic reticulum and FAD7/FAD8 in the plastid [18].

The changes in oil composition of edible seeds, especially the oleic and linoleic acid content can be modified genetically by silencing the FAD genes. High level of oleic acid in oil is one of the favored traits in oil engineering due to its high stability and several applications. Mutations in FAD2-1 genes have been reported to increase seed oleic acid content [19].

A rapeseed plant is genetically modified to produce oil with the desired balance of medium chain fatty acids and monounsaturated fatty acids. The activity of desaturases is reduced by introducing a stop codon into the coding sequence of the FAD2 gene to reduce the level of linolenic acid and linoleic acid, as well as to increase the level of oleic acid. The activity of the 9-stearoyl-acyl-ACP desaturase gene is modified with the *Macadamia integrifofia* gene to increase palmitoleic acid levels.

Modern DNA transfer technologies could ensure the transfer of genes that control the synthesis of low molecular weight triacylglycerols from species such as *Cuphea* to other, more traditional oilseeds. The chain lengths of fatty acids are determined by acyl-ACP thioesterases, which release the fatty acid chain from the fatty acid synthase. The activity of ketoacyl ACP synthase is reduced using the introduction of a stop codon in the coding sequence. The substrate specificity of palmitoyl thioesterase is altered by increasing levels of short- and medium-chain fatty acids by transforming the Cuphea gene with specificity for short chain lengths [20].

Esters with more branched carbon chain can also improve the low-temperature properties of biodiesel.

Thus, a plant (for example, rapeseed) can be genetically modified so that it produces an oil with triacylglycerol molecules containing residues of saturated acids with an average chain length and acids with 16-18 carbon atoms with one double bond in the radical (for example, palmitoooleic and oleic acid). Polyunsaturated fatty acid residues (as linoleic, linolenic and arachidonic) should be absent or present in a small amount.

Genetically modified rapeseed produces oil with a specific fatty acid composition comparing to unmodified ones (Table 4):

Table 4 – The content of methyl esters of fatty acids in biodiesel synthesized from genetically modified and unmodified rapeseed oil

Ester	Content of esters in samples synthesized from rapeseed oil, %	
	unmodified	modified
$C_7H_{15}COOCH_3$	–	5
$C_9H_{19}COOCH_3$	–	5
$C_{11}H_{23}COOCH_3$	–	15
$C_{13}H_{27}COOCH_3$	0,33	<1
$C_{15}H_{31}COOCH_3$	3,99	<1
$C_{17}H_{35}COOCH_3$	–	<1
$C_{15}H_{29}COOCH_3$ and $C_{17}H_{33}COOCH_3$	79,82	70
$C_{17}H_{33}COOCH_3$	2,09	<1
$C_{17}H_{31}COOCH_3$	6,03	<1
$C_{19}H_{39}COOCH_3$	4,32	
$C_{21}H_{41}COOCH_3$	4,60	
others		

The pour point of biodiesel produced from modified rapeseed oil is significantly lower (–20 °C comparing to –8 °C for unmodified rapeseed).

The more promising direction for producing of high-quality biodiesel is the use of oils obtained from the processing of microalgae.

Algae species capable of accumulation large amount of lipids are found in many taxonomic groups. However, *Chlorophytae* represent the biggest group within which the species with the average content of 25.5% of lipids in dry biomass have been identified [3].

The cultivation of microalgae can be organized in photobioreactors that provide a controlled environment (composition of the nutrient medium, temperature, illumination), which will make it possible to obtain oil with a constant fatty acid composition, independent from the growing area and climatic factors.

The algae are potentially more efficient in oil production in comparison to common oil seed crops due to the higher productiveness per area [21].

Genetic modification is part of the strategy to increase lipid production with algae. Target genes are lipid biosynthetic genes, lipid storage genes and lipid degradation genes. It is possible to alter lipid content by these genes modification. Another interesting aspect is the modification of the lipid characteristics. This could increase the quality of the lipids with regards to suitability as diesel fuel feedstock. The target genes are genes of fatty acid modifying enzymes, such as desaturases and thioesterases [22], [23].

Another possible approach to increasing the cellular lipid content is blocking metabolic pathways that lead to the such as accumulation of energy-rich storage compounds, starch. It was shown that starchless algae mutants accumulate increased levels of triacylglycerols during nitrogen deprivation [22].

4. Conclusion

The need to protect the environment makes it more common to use biodiesel instead of petroleum products. Vegetable oil products are preferred for producing of biofuels. Since the fatty acid composition of vegetable oil esters can vary, the physicochemical characteristics of biodiesel can also vary in a certain range. For a number of parameters, they do not meet the requirements of European standards.

Experimentally it has been shown that there is a correlation between the fatty acid composition and the physicochemical characteristics of biodiesel. The density, viscosity and low temperature characteristics of biodiesel depend on the ratio of medium and long chain fatty acid radicals, as well as on the degree of unsaturation.

Saturated unbranched radicals in the composition of biodiesel molecules worsen its low-temperature properties, but increase the oxidative stability and cetane number of biodiesel. The presence of double bonds improves the low-temperature properties of biodiesel, but reduces its oxidative stability. Thus, the presence of high contents of monounsaturated acids, as oleic or palmitoleic acids in biofuel is mostly preferred. The fatty acid composition of biodiesel can be changed by adding separately synthesized low molecular weight esters [24], [25]

Another way to change the composition of triacylglycerols in vegetable oil is modifying of oil-producing organisms (plants or microalgae) by genetic engineering methods. The target genes are those that determine the degree of unsaturation or the chain length of fatty acids.

Conflict of Interest

None declared.

Конфликт интересов

Не указан.

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