EFFECT OF BIODIESEL ON DIESEL ENGINE EMISSIONS

by

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Original scientific paper
https://doi.org/10.2298/TSCI18S5483N

Extensive research has been carried out with regard to the composition of the exhaust gases of Diesel engines in operation with biodiesel in relation to the operation with the conventional diesel fuel. Producing biodiesel from different raw materials and different technological biodiesel production processes can result in different individual physical and chemical characteristics of fuel. Generally, it can be said that the use of biodiesel (and mixtures) reduces the overall toxicity of the exhaust gases in relation to the operation of the engine with diesel fuel, and this is a significant environmental potential of biodiesel as a fuel for Diesel engines. However, there is a diversity of research results, due to different factors. The paper reviews and summarizes the relevant literature on the mentioned research that can contribute to the explanation of these effects. It also points to the need for a very careful selection of biodiesel for use as a Diesel engine fuel.

Key words: *Diesel engine, biodiesel, emissions*

Introduction

Observed from a wider perspective, the requirements set before alternative fuels for internal combustion engines can be classified into several general ones:

– the quality of an alternative fuel – the extent to which the characteristics of an alternative fuel are in accordance with the prescribed standards,
– the availability and renewability of raw materials for the production of an alternative fuel,
– the appropriate price of the cultivation of raw materials and the acceptable price of an alternative fuel on the market, and
– the fulfilment of ecological standards.

From the perspective of the usage of vegetable oil, or a fuel obtained from vegetable oil, as a liquid fuel alternative to the conventional diesel fuel, the previous requirements can be observed through the possibility of an alternative liquid fuel having certain advantages or at least no significant disadvantages compared with the conventional diesel fuel.

Generally, biofuels can be classified generationally, tab. 1, on the basis of the their production technologies and development sustainability including all of the implemented influences and effects from cultivation to even after exploitation [1-3]. The first generation of biofuels (often called *conventional biofuels*) are usually made from a raw material base used for food. The increase in the use of these raw materials for the production of biofuels has

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caused concern of various institutions, companies and researchers regarding the sustainability and global impact of the production of biofuels from the crops that are otherwise used for food and their general application. A number of analyses have been conducted in that sense, often with completely opposite conclusions. Furthermore, according to the authors of these papers, although the funding sources of these analyses and studies are not disclosed by rule, the impact of global and local energy conglomerates, companies and other spheres of influence and power on said analyses, studies and conclusions is enormous. The second generation of biofuels was meant to eliminate this potential effect and allow for the production of biofuels mostly from the crops that are not primarily used for food.

Table 1. Generational classification of biofuels [1-3]

Biofuel generation	Raw materials	Processing procedure	Fuel examples
First	Sugarcane, oil and other crops also used for food, animal fats	Fermentation, esterification	Bioalcohols, vegetable oil, biodiesel, biosyngas, biogas
Second	Crops not used for food, wheat, corn and similar vegetation, wood, waste	Fermentation, esterification, gasification, pyrolysis, etc., thermochemical processes	Bioalcohols, vegetable oil, biodiesel, bio-DMF, biohydrogen, bio-Fischer-Tropsch diesel
Third	Fast-growing crops and crops cultivated on *barren soil*, algae, microorganisms, crops developed by molecular biology techniques	Same as with the 2nd generation, molecular biology techniques	Bioalcohols, vegetable oil, biogas, biohydrogen, biodiesel
Fourth	Same as with the 3rd generation and crops developed by genetic engineering	Same as with the 3rd generation, genetic engineering, all with the CCS effect	Same as with the 3rd generation, all within the BECS concept

Generally, the third generation of biofuels encompasses biofuels obtained from fast-growing crops and crops cultivated on the soil that is not otherwise used for the cultivation of crops used for food, algae, microorganisms, and other crops and organisms developed by molecular biology techniques. The fourth generation of biofuels also includes genetic engineering in the development and processing procedures of raw materials, which enables the carbon capture and storage (CCS) of hydrogen compounds that are formed at the levels of raw materials cultivation (algae and microorganisms) and processing technologies (and would otherwise be emitted into the atmosphere) and their geological storage (*e. g.* in exploited gas and oil fields) or mineral storage (as carbonates). The bioenergy obtained from the fourth generation biofuels is said to satisfy the bioenergy with carbon storage (BECS) concept.

It is clear that the requirements (type of raw material, processing procedures) of the third, and especially the fourth generation of biofuels, reduce the number of countries that could potentially produce biofuels to technically and technologically developed countries, which are either not too dependent on imported conventional crude oil energy products or control this dependence by their economic, political and often even military power. Moreover, one should not forget that one of the primary initial goals of the application of alternative sources of energy (including biofuels) was precisely to reduce the dependence on fossil fuels.

Biodiesel as a fuel for Diesel engines – global advantages

As already mentioned, extensive research has been performed worldwide into the use of vegetable oils and their methyl and ethyl esters as fuels for Diesel engines, and from various perspectives at that. In the beginning, it was typical for the intensity of this research to grow with the occurrence of energy crises, shortages in fossil fuels (in the sense of a discrepancy between supply and demand) and rises in the price of crude oil (regardless of the reasons behind them), as well as the unstable forecasts on the real reserves of fossil fuels. Globally speaking, with the development of environmental awareness, production and processing technologies and techniques, the impulse of this research increased, with the use of biodiesel (and other alternative fuels) increasing as well, mainly due to the set regulatory requirements in relation to the minimal total participation of alternative energy sources (EU countries example, fig. 1).

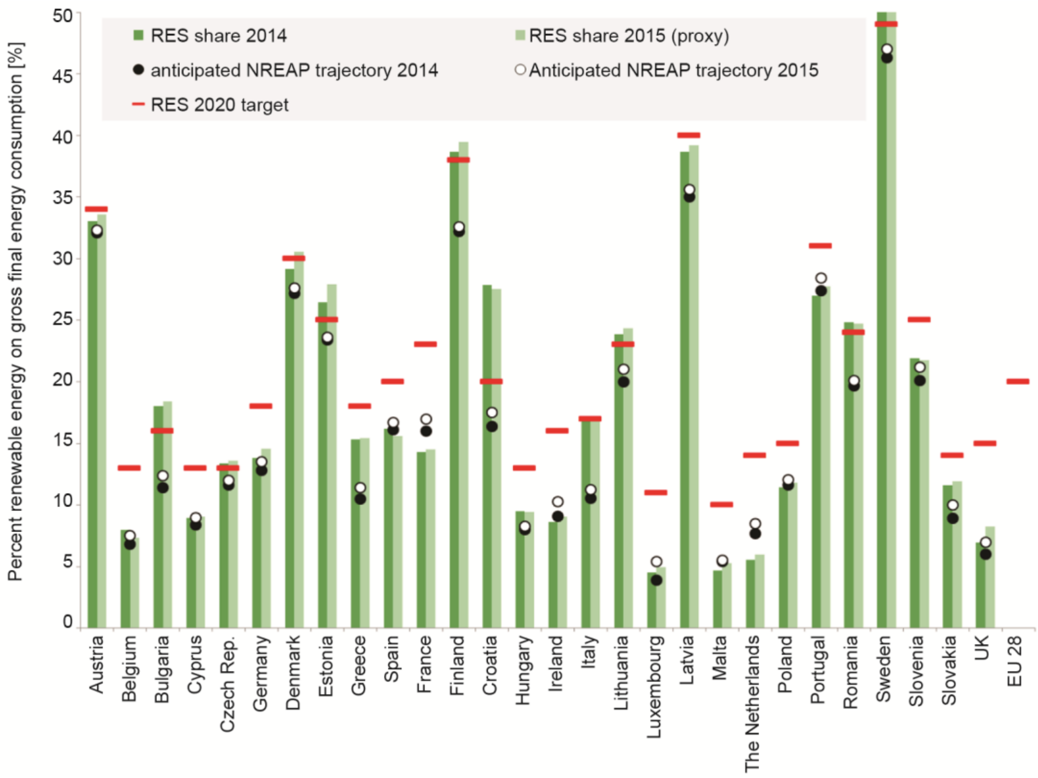

Figure 1. Member states current progress towards their 2013/2014 and 2015/2016 indicative red targets; National renewable energy action plan, data from member states (NREAP), RES; (source: Oko-Institut, EUROSTAT)

The basic advantages of using biodiesel compared with fossil diesel fuel can be observed in the following aspects.

– *Strategic* – biodiesel is a renewable fuel, with very similar characteristics to diesel fuel. To cultivate the raw material base of biodiesel it is not necessary to use the soil of a specific quality, the mid-quality, and even low-quality soil will do. The economic and the political stability of every country is inversely proportional to the importing dependence of a country with regard to fossil fuels and energy products, in general.

– *Economic* – the increase in the production of the raw material base for obtaining biodiesel, increases employment and the percentage of tillable soil of medium and low quality. The growth of employment is also reflected in the employment of people in the raw material to biodiesel processing plants as well as the development and expansion of the biodiesel distribution network. One should not neglect the effect of manufacturing the components of processing plants by the metallurgy and other industrial complexes.
– *Ecological* – which lies in the possibility for the reduction in the total emission of exhaust gases. Based on the *Ames mutagenicity tests* (USA), the use of biodiesel ensures the reduction in the various cancer hazards in people for up to 90%, compared with the risk carried by the use of diesel fuel, precisely due to the differences in the composition of exhaust gases [4]. The research [5] conducted with the aim of analyzing and finding the possibility for the reduction in air pollution caused by the passenger and freight vehicles, corroborated the fact that air pollution plays a significant role in people's health. Air pollution is represented as an important factor and cause of asthma, cancer, heart diseases, heart attacks, problems in heart functions, high blood pressure, appearance of congenital defects, and brain damage. The increase in instances of asthma has been recorded in the USA, while the direct presence of exhaust gases components from vehicles has been found in patients suffering from asthma, both in adults and children. The presence of air pollutants has also been confirmed in the people who have died suddenly, regardless of the age or sex of the population. The research has also proven that the influence of air pollution on the appearance of said diseases is particularly great in the vicinity of highly congested roads.

Biodiesel properties

The chemical procedure of esterification of vegetable oil with the aim of producing biodiesel is not a complicated process, and it basically adds up to breaking down large and complex molecules into simpler, and in size smaller ones. The fundamental explanation of this process is as follows: under the influence of potassium hydroxide (KOH) one mole of triglyceride (oil/fat), and three moles of alcohol are reduced to three moles of a mono-alkyl ester and one mole of glycerol with the residue of reaction products [6]. For example, in the reaction of 100 kg of rapeseed oil and 11 kg of methanol initialized by the activity of the catalyst (KOH – *caustic potash*), one can obtain 100 kg of rapeseed oil methyl ester and 11 kg of glycerol. Technological advancements have enabled the effect of saponification – the production of soap that is separated along with glycerol, to be bypassed so as to make possible the use of the obtained glycerol in the chemical industry.

If the base oil is, for instance, rapeseed oil, esterification with methyl ester yields the fuel called rapeseed oil methyl ester (MER, RME, ROME, FAME, or fatty acid methyl esters), while esterification with ethyl ester yields the fuel called rapeseed oil ethyl ester (EER, REE, ROEE, FAEE) and the like. Analogous to this, the biodiesel produced from soybean oil is labeled SME or SEE, *etc*.

Due to its characteristics, which are very similar to the characteristics of the conventional diesel fuel, this fuel has been named *biodiesel*, a label that clearly emphasizes the association with the conventional diesel fuel, while stating the fuel origin in its prefix.

When talking about biodiesel as a fuel for Diesel engines, the requirements concerning quality and characteristics that such fuels have to meet are defined by standards. In the USA this standard is ASTM D 6751, while in the EU it is EN 14214. In the Republic of Serbia the current standard is SRPS EN 14214, tab. 2, which is identical to the corresponding EU standard.

Table 2. Biodiesel characteristics according to the SRPS EN 14214:2012 standard

Property	Unit	Test method	Limits	
			min	max
Density at 15 °C	[kgm^{-3}]	EN ISO 3675 EN ISO 12185	860	900
Viscosity at 40 °C	[mm^{2}s^{-2}]	EN ISO 3104	3.5	5.0
FAME content	[%, mm^{-1}]	EN 14103	96.5	
Flash point	[°C]	EN ISO 2719 EN ISO 3679	101	
Cold filter plugging point (CFPP)[a)]	[°C]	EN 116		+5 summer –15 (–20) winter
Sulfur content	[mgkg^{-1}]	EN ISO 20846 EN ISO 20884 EN ISO 13023		10
Cetane number		EN ISO 5165	51	
Sulfated ash content	[%, mm^{-1}]	ISO 3987		0.02
Water content	[mgkg^{-1}]	EN ISO12937		500
Total contamination	[mgkg^{-1}]	EN 12662		24
Copper strip corrosion (3 hours at 50 °C)	Rating	EN ISO 2160	class 1	
Acid value	[mgKOHg^{-1}]	EN 14104		0.5
Linolenic acid methyl ester	[%, mm^{-1}]	EN 14103		12
Polyunsaturated methyl esters (≥ 4 double bonds)	[%, mm^{-1}]	EN 15779		1
Group I metals (Na + K)	[mgkg^{-1}]	EN 14108 EN 14109 EN 14538		5
Group II metals (Ca + Mg)	[mgkg^{-1}]	EN 14538		5
Methanol content	[%, mm^{-1}]	EN 14110		0.2
Monoglyceride content Diglyceride content Triglyceride content	[%, mm^{-1}]	EN 14105		0.7 0.2 0.2
Free glycerol	[%, mm^{-1}]	EN 14105 EN 14106		0.02
Total glycerol	[%, mm^{-1}]	EN 14105		0.25
Phosphorus content	[mgkg^{-1}]	EN 14107 FprEN 16294		4
Iodine value	[mg Iodine/100 g]	EN 14111 EN 16300		120
Oxidation stability (at 110 °C)	[hour]	EN 14112 EN 15751	8.0	

[a)] Climate dependent requirements

According to [6] the chemical formula of rapeseed oil methyl ester (MER) is $C_{19}H_{35.2}O_2$ with its molar mass of around 296 g/mol, while, for example, according to [7] the chemical formula of MER is $C_{21}H_{38}O_2$ with its molar mass of 323.4 g/mol, which is the consequence of the difference both in the basic oil and in the process of esterification itself. In relation to the basic oil, the molar mass of rapeseed oil methyl ester is approximately three times smaller [8]. The essence of esterification of rapeseed oil lies in the breaking down of large and complex molecules of oil into simpler and in size smaller ones. It is precisely due to the importance of the degree of esterification that this value is prescribed by a standard – according to SRPS EN 14214, tab. 2, the degree of esterification should be at least 96.5%.

The kinematic viscosity of biodiesel is somewhat larger than that of diesel fuel but it is significantly smaller than for basic oil, which is also directly related to the degree of esterification.

By comparing the distillation curves of crude oil and methyl ester [6] it can be concluded that for biodiesel the volatility curve is much more unfavorable compared with diesel fuel. From the perspective of application in Diesel engines, it would be desirable that volatility is higher in the region of lower temperatures. Namely, 50% of biodiesel volatility is achieved at the temperature that is around 80 °C higher that the respective diesel fuel temperature. This means that for the same mixture homogeneity of both fuels, biodiesel would theoretically require that much higher temperature in the compression chamber of the cylinder, which does not represent a problem in principle, bearing in mind that modern Diesel engines exhibit much higher temperatures than this.

Certain comparative characteristics of diesel fuels and biodiesels obtained from various basic oils are shown in tab. 3.

Appropriate standards prescribe the same minimal values of the cetane number for both fuels. Furthermore, an increase in the degree of esterification causes an increase in the cetane number [6, 31]. As necessary, the addition of certain additives can increase the cetane number of biodiesel.

As a qualitatively significant fact about biodiesel as a Diesel engine fuel, one can single out the mass ratio of oxygen at around 10% [32], *i. e.* 11.15% [15], as well as the lack of sulfates [4]. The presence of oxygen in the composition of biodiesel is important from the perspective of combustion. As far as sulfates are concerned, it is a fact that many studies show that they do not exist both in the composition of biodiesel and in the exhaust gases. However, certain studies [6] have confirmed the presence of sulfur oxides in exhaust gases, which can be explained by its origin from the soil in which the raw material base was cultivated. The presence of sulfates in biodiesel is predicted by appropriate standards, tab. 2.

Rapeseed oil biodiesel, compared with other biodiesels obtained from other vegetable oils, has proven to be one of the most convenient from the standpoint of usage as a Diesel engine fuel [6]. The majority of other biodiesels possess lower oxidation and thermal stability with an inadequate iodine value or inadequate filterability (CFPP) and cloud points (CP), tab. 3. Exceptionally, biodiesel obtained from algae occupies a very high position as a fuel for diesel engines according to its characteristics, tab. 3, yet as a relatively new fuel, with a specific production technology, it is still much less produced and used compared with biodiesels made from rapeseed oil, soybean oil, oil palms, *etc.*

The iodine value is an indicator of stability of biodiesel against oxidation. A biodiesel with a higher iodine value oxidizes more easily in contact with air. Also, a biodiesel with a higher iodine value tends to polymerize and form residue in injectors and piston rings. The iodine value depends on the raw material from which biodiesel is produced and it is limited by various standards in different parts of the world – in the EU (EN 14214, tab. 2) and Japan it is maximally up to 120 (in the EU even up to 130 for biodiesel as fuel oil), up to 140 in South Africa, not limited in Brazil, while in the USA, Australia and India it is not even included in the standards (so as not to exclude the raw materials such as soybean and sunflower oil from the production of biodiesel).

Table 3. Comparative characteristics of diesel fuels and certain biodiesels

Property	Diesel [a]	Biodiesel from oil of:							
		Rapeseed	Soya	Sunflower	Palm	Jatropha	Karanja	Mustard	Algae
Density (15 °C) $[kgm^{-3}]$	820-845	869-902[b]	876-925[b]	850[b]-884[u]	859[k]-883[b]	865[q]-882[e]	~894[g]	865[k]	820-890[x]
Viscosity (40 °C) $[mm^2s^{-1}]$	2-4.5	4.4[b]-5.65[u]	4.1[h], 4.4-4.9[b]	4.03[u]-4.9[b] 4.98[w]	3.7[u], 4.8[b]-5.7[h]	4.84[c]-5.56[u]	4.41[o] - 5.8[g]	5.66[u]-5.76[k]	3.68 [x]-4.52[u]
Flash point [°C]	min 56	166-179[u]	171[u] - 195[b]	89[w] -187[b]	167[b] - 176[u]	170[u] - 191[c]	114[r] - 168[o]	150[k]	>160[u]
Cetane number	min 51	>51[m] - 54[r]	48[b] - 51.3[p]	49[b]	59[k]- 64.6[j]	51-52[c]	50.8[o] - 54.5[r]	54.9[u]	51-65.5[x]
Lower heating value $[MJkg^{-1}]$	~42.5[d]	36.3[b] - 38.2[m]	36.7[b] - 38.4[f]	~36[b] - 38.4[u]	36.3[k] - 37.5[u]	38.5[c]	35.9[r] - 37.9[o]	~36.7[k]	33.3-36.5[x]
Cold filter plugging point CFPP [°C]	max +5 summer max -15 winter	-10 to -6[u], -9[s]	-7 to -5[b], -2[s]	-4[u] to -12[w]	10 to14[u], 12[s]	2[s]	3[t]	-5[u]	-2.6 to -11.7[x]
Cloud point CP [°C]	max +3 summer max -5 winter	-3[s], -1 to 8[b]	0 to 1[b]	-1[b], 0 to 4[u]	6 to 8[b], 16[k]	4[s], 13[q]	6[w] - 13.6[g]	4[u], 5[k]	-5[u]
Iodine value [mgJ per 100 g]	-	97.4[u] - 114[v]	120-133[u]	132[n]	50[v] - 59[u]	105[u]	83[o]	102[k]	65-109[x]
Oxidation stability (110 °C) [hour]	max 2.5[y]	6.5[v]	7.1[l]	0.8[u]-2.7[w]	14.7[v]	2.3[u]	2.98[w]	1.1[u] -16[k]	5.6-95.7[x]

[a] EN 590, [b] [9], [c] [10], [d] [6], [e] [11], [f] [12], [g] [13], [h] [14], [j] [15], [k] [16], [l] [17], [m] [18], [n] [19], [o] [20], [p] [21], [q] [22], [r] [23], [s] [24], [t] [25], [u] [26], [v] [27], [w] [28], [x] boundary values for 9 different algae with cetane number ≥51 and CFPP <0°C [29], [y] in mg/100 ml acc. EN 590, [z] [30].

Engine performance

In principle, biodiesel combustion characteristics are very similar to the conventional diesel fuel combustion, leaving no significant coke residue at that. The use of biodiesel as a Diesel engine fuel may be in the form of pure biodiesel (B100) or a mixture of biodiesel and classical diesel fuel in various mix ratios (most commonly B5, B7, B10, B20, *etc*.). The B20 marks the fuel mixture of biodiesel and diesel fuel with 20% biodiesel and 80% diesel, and so on. Biodiesel and diesel fuel mixtures, as well as pure biodiesel, can be supplemented by various substances with the aim of enhancing fuel characteristics and engine operation.

When using biodiesel and biodiesel/diesel mixtures, one can observe a drop in the value of the effective engine power and effective engine torque compared with the operation with diesel fuel [33]. This value decreases with an increase in the ratio of biodiesel in the mixture with diesel. The greatest drop was observed in B100 fuel in the engine operation modes that correspond to the maximum (nominal) effective power and maximum effective torque (5-10%). The authors explain this by a lower bottom thermal power of biodiesel (RME) compared with diesel fuel. Furthermore, the effective specific fuel consumption also increases by maximally about 12% for B100 fuel. During the use of a mixture of soybean biodiesel (SME) and diesel fuel in the identical volumes (B50), the authors [34] did not observe any significant change in the engine power, however, they point to an increase in the specific fuel consumption, also due to the differences in the bottom thermal power of fuel and higher density and viscosity of B50 compared with diesel fuel. Ozener *et al.* [17] reached similar conclusions in their research results, which showed a drop in the effective engine torque of about 1.5% (for B10) up to 4.7% (for B100), with an increase in the effective specific fuel consumption from around 2% to around 9% (respective of the increasing ratio of biodiesel in the mixture), in relation to the engine operation with diesel fuel, due to the lower bottom thermal power of biodiesel compared with diesel. The authors also observed a drop in the temperature of exhaust gases from about 1.46% to 5% (with the increasing ratio of biodiesel in the mixture). During the use of the mixture of Karanja biodiesel and diesel, the drop in the effective torque becomes evident only with the ratios of biodiesel in the mixture of over 20% [13]. Experimental research into the Diesel engine performance when operating with Mustard biodiesel [16] and coconut oil biodiesel mixtures with diesel fuel [35] also points to a small drop in the engine power and torque, with an increase in the specific fuel consumption, due to the lower bottom thermal power and higher density and viscosity of biodiesel compared with diesel. In the review paper [36], it is emphasized that the majority of papers show that the drop in the engine power when operating with B100 compared with diesel fuel is smaller in percentage terms, while the increase in the specific fuel consumption is approximately in the range of the percentage differences of thermal powers of fuels. Also noted is a somewhat higher hourly consumption of the B100 fuel compared with the engine operating with diesel. The lower bottom thermal power, higher viscosity and density, followed by inferior dispersion and larger fuel drops and lesser quality of mixture formation, are all causes of a slight drop in the power and degree of efficiency along with an increase in the specific fuel consumption [37]. The analysis of the characteristics of fuel injection points to the earlier start of injection, longer duration, and an increase in the pressure of injection, with an increase in the ratio of biodiesel in the mixture with diesel fuel [38]. Due to the lower bottom thermal power of B100, the effective power is smaller for about 5%, the effective specific consumption is larger for about 10%, while the temperature of exhaust gases and pressures in cylinders are lower than when the engine operates with diesel fuel. However, it is possible to find the optimal pre-injection time so that the effective power, effective specific fuel consumption, exhaust gases temperature, pressures in cylinders and other important characteristics of engine operation can be at an acceptable level when using B100 compared with diesel [39]. The research [8, 14, 31, 40-46] shows that from the aspect of using biodiesel as a Diesel engine fuel and the effects on engine operation, it is important that processes that take place in the fuel supply systems, injection and mixture formation processes, as well as knowledge of biodiesel characteristics that are not prescribed by standards (speed of sound, bulk modulus, surface stress, *etc.*) are fully studied and clear.

Emission characteristics

Numerous studies have been done on the comparison of the composition of exhaust gases from Diesel engines operating with biodiesel or biodiesel/diesel mixtures with engines operating with the conventional diesel fuel.

The research conducted in this direction [13, 15, 17, 18, 34, 36, 37, 39, 42, 43, 47-53] undoubtedly points to the important smoke reduction in exhaust gases when using biodiesel compared with diesel fuel. The reduction is mentioned as *significant* in all of the conclusions of the previous studies and it ranges from 35% [37] to 72% [49] when using B100 compared with diesel fuel. The studies [36, 37, 49, 50] are reviews and they present the conclusions of a larger group of researchers. When using a fuel with a different mixture composition of biodiesel and diesel fuel, one can observe an increasing trend in the smoke reduction with an increasing ratio of biodiesel in the mixture. Thus, for example, in [49] the smoke reduction is from 5.5% for B20 to 35% for B100 compared with diesel fuel, while in [17] the smoke reduction in the engine operating with B10, B20, B50, B100 is 19.5%, 26%, 45.5%, and 64.2%, respectively. Li *et al.* [53] achieved a reduction of smoke of around 52% when using B30 compared with the engine operating with diesel fuel. When using B100 obtained from palm oil, in the operation of a DI Diesel engine (with the high pressure common rail system), a turbocharger with variable geometry and an exhaust gas recirculation (EGR) system, the authors [48] observed a smoke reduction in all modes of operation, while when the same engine operated with B20 there appeared an *isolated case* of slight increase in smoke with the reduction in NO_x concentration, compared with the engine operating with diesel fuel.

In accordance with the smoke reduction, also significantly reduced is the emission of particulate matter (PM) in the exhaust gases when using biodiesel compared with diesel fuel [18, 32, 36, 38, 49-51, 54, 55]. The reduction in particles reaches even 70% [55], while the review paper [49] states that the reduction in particles is in the range from 50% to 69%, and even shows the extreme cases of 75% to 91% when using B100 compared with diesel fuel.

The reduction of CO, when using biodiesel and Bxx mixtures compared with diesel fuel, is also significant [13, 16-18, 21, 32, 33, 35-37, 39, 42-44, 47, 49, 50, 52, 54] and it ranges from 19% for B10 (10% mustard biodiesel and 90% diesel) [16], 25% for B100 (rapeseed oil biodiesel) [18, 39], 20-50% (biodiesels of different origin) [37], 70% for B100 (Karanja biodiesel), up to the extreme 73-94% for B100 (karanja biodiesel) [49]. When using the B30 fuel, in lower engine operation modes, one can observe a slight increase of CO emission, while in the higher operation modes the reduction of around 43% CO is noticeable in comparison with the engine running on diesel fuel [53]. Sadeghinezhad *et al.* [49], in their research state that around 84% of researchers, whose papers they reviewed, undoubtedly conclude that when using biodiesel (or its mixtures) the CO reduction is significant compared with the use of diesel fuel. In the remaining 16%, there are examples of an increase in CO emission when an engine operates with biodiesel compared with diesel fuel, in the range from 6.7% to 37%.

The CO_2 emission is a particularly interesting research topic, not only in the context of using biodiesel as a fuel for Diesel engines, but in the wider scope of things, the so-called indirect land use change impacts of biofuels, also known as ILUC. This relates to the consequences of changes in soil purpose worldwide due to the increased crop surfaces used for the production of ethanol or biodiesel and the emission of a greater amount of CO_2 (and carbon in general from the processes of biofuels production), as a reply to the increased global demand for biofuels.

The studies [56, 57] on the influence of biodiesel application to the greenhouse effect at the global level point to the possibilities of the total reduction in CO_2 by 50-80%. With regard to CO_2 emission when biodiesel is used as fuel, the National Biodiesel Board (NBB, USA) [4] recommendation on using biodiesel as a substitute for diesel fuel states the following data: the composition of exhaust gases from biodiesel combustion, compared with diesel fuel, possesses the reduced presence of CO_2 of up to 80%, sulfates up to 100%, total unburned hydrocarbons (HC) over 90%, while the reduction in aromatic HC is between 75% and 90%, with a substantial decrease in the emission of PM and CO. Substituting a tonne of diesel fuel with a tonne of biodiesel results in the reduction of around three tonnes of CO_2 in the atmosphere [58]. The increased presence of CO_2 in the composition of exhaust gases from an engine operating with B100 compared with diesel fuel has been observed by authors [42]. When an engine operates with B5 and B15 mixtures (coconut oil biodiesel) the author [35] states that the increase in CO_2 is from 2.5% to 4.6%. Comparing the emissions of exhaust gases from the engines using B5, B20, B50, and B100 (soybean oil), the authors [17] state that the increase in CO_2 is 1%, 1%, 2.8%, and 5.6%, respectively, when compared with the engine running on diesel fuel. The increase of biodiesel (with characteristics according to SRPS EN 14214) in the blend leads to the reduction of CO_2 and CO emissions [59]. With higher loads CO_2 emission is reduced less than the CO emission. This is the result of more complete fuel combustion at higher engine load, authors conclude [59].

In the majority of papers, there is a significant reduction in unburned HC, when diesel fuel is substituted with biodiesel and its mixtures [13, 16-18, 35-37, 42, 49, 50, 52-54]. The reduction of HC ranges from around 30% [18], to 42% [16], to 55% [37], and to 66% [13]. When using B10, B20, B50 mixtures and pure B100 biodiesel (soybean), the reduction of HC is 20%, 23%, 31%, and 44%, respectively [17]. Sadeghinezhad *et al.* [49], in their review paper, state that the reduction of HC can be even up to 89% and provide an isolated example of a decrease in HC emission of around 10% noticed (jatropha biodiesel) when biodiesel was used instead for diesel fuel.

The research results related to the emission of NO_x vary to a certain degree. For different engines and different testing conditions, biodiesel of various origin and different biodiesel/diesel mixtures, NO_x emission can be lower for around 20% but also higher for around 26% [49], or as in [37] – lower for 26%, or higher for around 14% when using biodiesel instead of diesel fuel. The increase in NO_x emission is present in the research results in [35, 36, 42-45, 60], while the papers [21, 32, 38, 55] show the increase of NO_x emission of 5%, 10%, 15%, and 16 %, respectively, when biodiesel is substituted for diesel fuel. When using B30, NO_x emission is higher in lower engine operation modes (around 8%) than in higher modes (around 1%), compared with the engine running on diesel fuel [53]. A similar conclusion is reached by Valentino *et al.* [34]. When using B100, B75 and B50, a drop in NO_x emission is observed, while this emission increases in the case of B25, particularly in higher operation modes, compared with the engine running on diesel fuel [33]. In the cases where biodiesel represents 15% of the total mixture with diesel fuel there are no effects on NO_x emission, while this emission increases with the increasing ratio of biodiesel in the mixture and it amounts to 20% for B20, and around 29% for B100, compared with the engine running on diesel fuel [13].

An increase of NO_x emission with an increase in the ratio of biodiesel in the mixture with diesel fuel is one of the conclusions of the research [17], thus the use of B10, B20, B50, and B100 increases NO_x emission by around 7%, 11%, 12%, and 18%, respectively. The authors in [50, 54] in their review papers conclude that the majority of studies (66.5% of studies [54]) show an increasing trend of NO_x emission when using biodiesel instead of diesel fuel. Deviations from the composition of the reference fuel for which an engine is intended and optimized may lead to the emission of exhaust gases exceeding permitted limits. This is a potential problem for the use of biodiesel in the future. It seems that the problems related to the emission of exhaust gases when using biodiesel first appeared with the introduction of stricter regulations on the emission of exhaust gases since the Euro III norms (in EU). This requires an engine to be adjusted to operating with biodiesel and certain biodiesel/diesel mixtures. Thanks to the EGR the reduction in NO_x emission is achieved with a small increase of smoke, when an engine operates using B20 compared with diesel fuel [48]. The reduction of NO_x emission achieved by optimizing the time of B100 pre-injection is even up to 25% compared with the engine running on diesel fuel [18, 39]. In a similar manner and with 17% EGR, the reduction of NO_x emission was also achieved without significant effects on CO and HC emission [52]. Adding certain substances to biodiesel allows for a reduction of NO_x emission compared with the operation using pure biodiesel – the addition of bioethanol [46], *i. e.* the addition of ethanol and n-butanol [47], is recognized by the authors as a possibility and potential for the reduction of NO_x emission. The reduction in NO_x emission of up to 9% in relation to the operation using diesel fuel was achieved by the authors [53] by adding pentanol to the B30 mixture. Adding the AdBlue additive to the exhaust gases before the SCR catalyst leads to a significant reduction of NO_x emission compared with the engine running on tested fuels (diesel, B7, B20, B50 and B100) without said additive [51]. The NO_x emission from the engine operating with all of the tested fuels was in accordance with the Euro IV norm, except for B100 where a somewhat higher NO_x emission than the permitted was noted.

Table 4. Boundary changes in the composition of exhaust gases when using biodiesel instead of Diesel fuel in diesel engines

Components in exhaust gases	For B100 compared with diesel	
	Reduction [%]	Increase [%]
Smoke	35 to 72	–
Particulate matter	50 to 70(90*)	–
Carbon monoxide	20 to 50(90*)	6 to 37
Total unburned hydrocarbons	44 to 66(89*)	to 10
Nitrogen oxides	to 26	to 29
Sulfates	to 100	–
Polycyclic aromatic hydrocarbons (PAH)	to 80	–
Nitrated PAH	to 90	–

* values in parenthesis are individual cases, extreme values

The use of the 20% water and 80% biodiesel emulsion resulted in the reduction of NO_x by 41% compared with the engine operating using B100 and around 30% compared with diesel fuel, with the simultaneous reduction in HC emission (3.5% less than B100), smog (9.6% less than B100), and a drop in the efficiency degree of around 4% (in relation to B100)

[61]. By using different test additives (among others, antioxidants – vitamins C and E), the group of authors [22] achieved the reduction of NO_x, however, with an increase in CO and HC emission. From the emission test it is found that B20 has better emission characteristics comparing to diesel except NO_x, the addition of leaf extract (LE) slightly reduces the NO_x emission of B20 and appreciably suppresses smoke emission level [62]. The measurements of harmful emissions at all modes of the emission test cycles (ESC) test show that the NO_x, CO, HC, and smoke emissions may generally decrease with higher content of biodiesel, if the injection pump timing is optimized [63].

On the basis of the previous analysis of the data from the literature related to the emission of exhaust gases, tab. 4. shows the boundary changes in the composition of exhaust gases when using biodiesel instead of diesel fuel.

Analysis of some of the papers that are referenced to the authors [49] for increasing the CO emissions (when an engine operates with biodiesel compared with diesel fuel), can be noticed the following.

- In four of the five references [64-67], were used biodiesels with characteristics not according to the EN 14214: 2012.
- Lujan *et al.* [64] did not mention the origin and characteristics of biodiesel.
- The physico-chemical properties of the jatropha, karanja, and polanga oil based biodiesel and their various blends with diesel were evaluated as per the ASTM standards [65] (it is known that, for example, the value of the iodine number is not included in the ASTM D6751 standard).
- Banapurmath *et al.* [66] used honge oil (HOME, known as karanja oil or pongamia pinnata oil), jatropha oil (JOME) and sesame oil (SOME) with no data about iodine value, FAME content, *etc.* (data for density, flash point, kinematic viscosity, and lower heating value are given from the characteristics of biodiesel).
- Kumar, *et al*. [67] used jatropha biodiesel with known properties like in [66], with no data about iodine value, FAME content, *etc*.

Sadeghinezhad [49] observed *an amazing trend represents the increase of HC emissions from biodiesel* [66, 68, 69]. In addition, for soybean biodiesel [68] where the most biodiesel properties were found to agree with the standard EN 14214 limits, except the iodine number which was found to be slightly higher. The increase of HC emission was about 10% from methylester of jatropha oil in comparison to diesel fuel [67]. Banapurmath *et al.* [66] have established that HC emissions with HOME, SOME, and JOME were higher in comparison to the standard diesel fuel in a one cylinder, 4-stroke, WC, DI engine at 1500 rpm. Kumar *et al.* [69] used jatropha biodiesel and mixtures (with methanol, hydrogen, and amazing one with orange oil) with incomplete data on biodiesel characteristics. Xue *et al.* [70] observed this trend in a relatively poor atomization and lower volatility status of biodiesels.

Only on the basis of the previous few studies, if we would exclude references, where biodiesel is used without any clear origin and characteristics or characteristics that do not according to the EN 14214 standard, from tab. 4, it would give a much more favorable estimate of the use of biodiesel, especially in terms of CO and HC emissions.

Conclusions

Generally speaking, the use of biodiesel (and its mixtures) reduces the total toxicity of exhaust gases compared with diesel fuel, which is a significant ecological potential of biodiesel as a fuel for Diesel engines. However, there is a dispersion in the obtained research results, which is the consequence of several factors.

- Producing biodiesel from different raw materials and different technological biodiesel production processes can result in different individual physical and chemical characteristics of fuel (even though it satisfies the regulatory standards).
- Regulatory standards for biodiesel are different in the world. This can lead to different conclusions even for the same engine when using biodiesel from different raw materials or in different parts of the world or the climates.
- Testing is performed on different types of Diesel engines produced by different companies, single or multi-cylinder engines, pre-chamber, chamber and DI engines, with various fuel supply systems and various (im)possibilities of regulating the operation of these engines, and other varying characteristics of engines on the whole (air supply systems, combustion space characteristics, existence and operation of devices for the treatment and re-circulation of exhaust gases, *etc*.).
- Testing is performed in different engine operation modes and for different ratios of biodiesel in the mixtures with diesel fuel, *etc*.

On the basis of the previous analysis, the prospects of the production and use of biodiesel as a fuel for Diesel engines are clear. The research has been intensified in all areas from the manner and technologies of cultivating (obtaining) the raw material base for the production of oil, biodiesel processing and production technologies, and finally to the application in diesel engines.

For the application of biodiesel as a fuel in Diesel engines, it is necessary to:

- align the biodiesel (and mixtures) characteristics with the appropriate standards and that there is a stable source of biodiesel with constant properties,
- fully study and clarify the processes that take place in the fuel supply systems, injection processes, mixture formation and biodiesel combustion, as well as emission characteristics and
- know the fuel characteristics that are not prescribed by standards (speed of sound, density, bulk modulus, surface stress, *etc.*), yet are very important from the perspective of their influence on the processes of injection, mixture formation, combustion and emission of exhaust gases.

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Paper submitted: April 8, 2018
Paper revised: July 7, 2018
Paper accepted: July 9, 2018