

THE EXPLOITATION AND ENVIRONMENTAL CHARACTERISTICS OF DIESEL FUEL CONTAINING RAPESEED BUTYL ESTERS

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Abstract. In an effort to consume fewer non-renewable resources and use primarily raw materials of biological origin in the production of biofuel, biomass-derived biobutanol can be used for transesterification of rapeseed oil. The study investigates the physical and chemical characteristics of rapeseed oil butyl esters (RBE) and 10%, 20% and 30% RBE-diesel composites. Characteristics of RBE and their composites with conventional diesel (D) are a little different from rapeseed oil methyl esters (RME) and their composites with D, while their basic characteristics comply with the requirements of European Standard EN 14214. Comparative tests in 4-stroke 4-cylinder 1Z type diesel engine of Audi-80 using 10–30% RBE and 10–30% RME compounds as fuels have been performed, and the results indicate that the biologically derived additives improved the energy characteristics of the engine but increased the fuel consumption compared to pure diesel. Increasing the bio-component concentration up to 30% in diesel and biodiesel fuel (RME and RBE) mixtures leads to complex improvements in the environmental impact compared to pure diesel; however, in the case of mixtures with RBE, slightly higher concentrations of CO₂, HC and NO_x were observed in engine exhaust gases compared to RME and D mixtures of analogical composition.

Keywords: biofuel; biodiesel; biobutanol; diesel engine; engine emissions.

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Introduction

Many anthropogenic activities, particularly automotive transportation, have contributed significantly to environmental pollution and greenhouse gas effect around the world. One way to reduce automotive emissions that contribute to the greenhouse effect and environmental pollution is using renewable energy sources. Biodiesel fuel made from raw materials of vegetal or animal origin by transesterification with alcohols (methanol, most commonly) can be used in diesel engines (Balat, M., Balat, H. 2010). The performance characteristics of biodiesel fuel resemble those of conventional diesel (D), and biodiesel provides the benefits of being derived from renewable raw materials and releasing less hazardous gases into the environment (Fazal *et al.* 2011). Results of the tests carried out have shown that combustion of biodiesel derived from rapeseed oil methyl esters (RME) generates lower levels of hazardous components (excluding nitrogen oxides, NO_x) in exhaust gases (Knothe *et al.* 1998; Benjumea *et al.* 2008).

Exhaust gases from combustion of D are composed of many components in gas or solid (particle) phase. Some of these components, such as formaldehyde, benzene and sulphates, are confirmed or suspected carcinogens. Burning D contributes to release of high levels of solid phase materials composed of metals, sulphates, soluble organic fractions and aromatic hydrocarbons (Maricq 2007; Ristovski *et al.* 2012). Results from epidemiological studies have shown that elevated levels of particulate matter ($d < 0.3 \mu\text{m}$) emitted in engine exhaust contribute to increased morbidity and mortality rates. Statistical analysis of these data suggests that sulphate levels correlate with general mortality rate, whereas organic fractions (e.g. poly-cyclic aromatic compounds) correlate with mutagenicity of diesel fuel emissions (Westerholm *et al.* 1991; Kleindienst *et al.* 1999). Studies investigating the long-term human health effects of diesel engine emissions have reported a link to lung cancer. Additionally, epidemiological studies have

found that the short-term and long-term health effects of particulate matter include cardiovascular disease and airway disorders, along with increased mortality (Hesterberg *et al.* 2006; Langrish *et al.* 2009).

Volatile Organic Compounds (VOCs) play a significant role in photochemical smog formation. VOCs react with NO_x in sunlight and form photo-oxidants when OH radicals are present in the atmosphere. Fuels are the main sources of VOC and NO_x emissions in urban areas. To reduce the health and environmental effects of automotive emissions, the use of alternative fuels has been investigated.

The operating characteristics of diesel engines using biodiesel fuel, particularly RME-derived diesels, have been extensively investigated. Detailed emission tests for diesel engines operating on RME have been performed (Mittelbach *et al.* 1985; Krahl *et al.* 1996). The tests have shown that the use of RME alone or in blends with D reduces smokiness and the concentration of sulphur oxides, aromatic hydrocarbons and CO; however, higher emissions of NO_x and soluble organic fractions have been observed. Schumacher *et al.* (1996) have found that, in the case of 100% biodiesel, NO_x levels increased by 13% and Particulate Matter (PM) decreased by 20% compared to exhaust emissions from pure D. Similar results have been found by Chang *et al.* (1996), who evaluated diesel mixtures containing soybean oil methyl- and isopropyl-esters. Conversely, research by Peterson *et al.* (1996) that was performed under identical conditions demonstrated that biodiesel causes a 12% decrease in NO_x but a 10% increase in smokiness when compared to D emissions.

Wang *et al.* (2000) have investigated exhaust gases from 35% RME-diesel mixtures and found a 12% reduction in CO levels and a 25% reduction in carbonate derivatives and health-hazardous particulate levels. This reduction was caused by factors such as higher levels of oxygen, higher cetane number and lower levels of aromatic carbohydrates and sulphur compounds. However, NO_x emissions are often dependent on the engine age, because the formation of NO_x largely depends on the gas chamber temperature and pressure.

With the goal of using renewable raw materials and reducing the detrimental impact on the environment, the use of bioalcohols (alcohols made of biomass) has increased. Biobutanol can be produced from biomass by the same companies that produce bioethanol because the manufacturing technologies of the two products are nearly similar. There is a particular interest in producing biobutanol because of its better performance compared to ethanol in Otto engines in mixtures with conventional gasoline. Several studies have analysed in detail the engine performance and environmental characteristics of gasoline–butanol fuel mixtures and even fuel burning kinetics. Pukalskas *et al.* (2009) found that increasing the butanol concentration to 50% in mixture with gasoline cuts the

emissions of CO and CO_2 in exhaust gases to 80% and 15%, respectively, of gasoline alone. No significant changes in CH concentrations were observed. It should be noted that fuel consumption increased due to the lower calorific value of butanol.

Diesel fuel consumption (compared to petrol) has been growing, and it is therefore reasonable to orient new types of raw materials towards diesel fuel production. Biobutanol could replace synthetic methanol used for transesterification of oil or fat in producing biodiesel. This application would improve the environmental characteristics of the fuel, and renewable raw materials would be used exclusively to produce the fuels.

Laza *et al.* (2006) analysed engine performance and fuel burning characteristics using butanol blends with diesel fuel as fuel for a diesel engine. Lebedevas *et al.* (2010) found that the use of tertiary mixtures containing D, butanol and fatty acid methyl- or butyl esters rather than D–butanol mixtures is more reasonable for diesel engines. Using fuels with 30% bio-components reduces the CO_2 concentration in exhaust gases by 20–25% and reduces smokiness by 5–7 times compared to D fuel alone. In these experiments, equal proportions of RME and RBE were used.

The aim of our work was to determine whether RBE and D mixtures could be used in automotive engines and to evaluate the environmental benefit of these fuel mixtures. To accomplish this goal, an analysis of the physical and chemical characteristics of the mixtures as well as an evaluation of engine performance and exhaust emissions was completed. In the light of current regulations that encourage the replacement of fossil fuels with biofuels, the authors chose higher concentrations (up to 30%) of RBE and D mixture than are widely used.

1. Materials and methods

The following materials were used for these experiments.

Refined rapeseed oil for synthesis obtained in a local market. The oil was stored at 4 °C.

n-Butanol, purity 99.5% (Chempur, Poland).

RME received from the biodiesel production company JSC ‘Mestilla’, and compliant to requirements of the European Standard EN 14214:2012. Esther level: 96.8%.

RBE obtained in laboratory conditions by applying the three-stage oil transesterification with butanol procedure using an alkaline catalyst (metal sodium). The RBE was synthesised in a thermostatic reactor with a hermetic mixer. The oil was homogenised by maintaining a constant temperature (+105 °C) and a constant mixing speed (300 min^{-1}), and butanol (in the molar proportion of 7:1 to oil mass) with dissolved catalyst (metal sodium, 0.3% of oil mass) was added. In stages 2 and 3, the molar proportion of

butanol and oil was 3:1, and the level of catalyst was 0.1% of oil mass. The duration of one stage of the synthesis was 2 hours. After the reaction was completed, the RBE was washed with a 10% phosphoric acid solution and distilled water. Residues of water and butanol were eliminated using a vacuum rotational evaporator. The RBE was purified by adding 2% silica gel (Sigma Aldrich) and then filtering. The ester content in RBE was 97%. The remaining characteristics of RBE were fully compliant with the European Standard EN 14214:2012.

D was obtained from a fuel marketing network. The quality of the diesel complied to the European Standard EN 590 Grade C.

The fuel compounds for testing were prepared by mixing the diesel (D), RME and RBE. Compounds in the following proportions were tested:

- 90% D and 10% RME (abbreviation 10% RME);
- 80% D and 20% RME (abbreviation 20% RME);
- 70% D and 30% RME (abbreviation 30% RME);
- 90% D and 10% RBE (abbreviation 10% RBE);
- 80% D and 20% RBE (abbreviation 20% RBE);
- 70% D and 30% RBE (abbreviation 30% RBE).

The viscosity and density of the compounds were measured with Stabinger viscometer Stabinger SV3000.

The calorific value was measured with the calorimeter IKA C200. The elementary composition of RBE, RME and their mixtures were evaluated with the CHNS/O analyser PerkinElmer 2400 Series II. The cetane number was calculated using Omega 1.5 software (Petroprogram Company). Cold Filter Plugging Point (CFPP) tests were carried out according to the requirements of the European Standard EN 116:1997.

The 4-stroke 4-cylinder 1Z type diesel engine of Audi-80 was chosen as a testing object. The engine had direct injection, a turbo compressor, an exhaust gas recirculation system and an Electronic Control System (ECS). ECS evaluates working regime of the engine, environmental air conditions and other related parameters and optimises the quantity of fuel and the

time of injection. There was an exhaust gas recirculation system designed to reduce pollution with NO_x . As the engine power grew, the ECS system reduced the exhaust gas recirculation.

For basic technical characteristics of the engine, see Table 1.

The testing was carried out in the automated engine test stand KI-5543 with a weighing dynamometer for evaluation of the torque of the internal combustion engine.

An electric asynchronous motor with phase rotor, fluid rheostat and dynamometer was used for engine brake on the engine test stand. The stand is universal at $600\div 1400 \text{ min}^{-1}$ revolution and operates as an electric motor; at $1600\div 3000 \text{ min}^{-1}$ revolutions, it operates as a power generator. The torque of the stand is controlled by an electromechanical drive fluid rheostat in both the motor and generator modes. The maximum braking torque of the stand is 186 Nm, and the maximum measurable torque is 440 Nm. The torque measurement bias is 0.8% of the maximum torque value.

The engine was directly (not through the gearbox) connected to the stand by means of a shaft with equal angular velocity hinges on both ends.

Fuel consumption was measured with an SK-5000 electronic scale, which had a maximum weighing capacity of 5000 g (one graduation value was 1 g). The accuracy of the hourly fuel consumption measurement was 0.5%. To eliminate the measurement error each test was repeated three times.

An AVL DiCom 4000 exhaust gas analyser was employed for the analysis of the hazardous components of the exhaust gas from the engine. For the exhaust gas measurement range and accuracy, see Table 2.

2. Results and discussions

2.1. Characteristics of fuel mixtures

The physical and chemical characteristics of the fuel are very important because they determine the engine performance and emissions quality. Fuel density and viscosity affect fuel injection. Fuel fluidity

Table 1. Parameters of tested diesel engine

Parameter	Value
Displacement (cm^3)	1896
Number of cylinders	4
Compression ratio	19.5
Power (kW)	66 (4000 min^{-1})
Torque (N m)	180 ($2000\div 2500 \text{ min}^{-1}$)
Mean effective pressure (MPa)	1.19 (180 N m)
Cylinder diameter (mm)	79.5
Piston stroke (mm)	95.5

Table 2. Measurement range and accuracy of gas analyser AVL DiCom 4000

	Measurement range	Resolution
Opacity	$0\div 100\%$	0.10%
Absorption (K -value)	$0\div 99.99 \text{ m}^{-1}$	0.01 m^{-1}
Nitric oxides	$0\div 5000 \text{ ppm (vol.)}$	1 ppm
Hydrocarbons	$0\div 20,000 \text{ ppm (vol.)}$	1 ppm
Carbon monoxide	$0\div 10\% \text{ (vol.)}$	0.01% (vol.)
Carbon dioxide	$0\div 20\% \text{ (vol.)}$	0.10% (vol.)

is important for exploitation. Fluidity defines the ability of the fuel to flow through the engine power system tubing and assembly and depends on the fuel viscosity and filtration characteristics. Fluidity is assessed according to indicators such as viscosity, clouding temperature (CP) and CFPP. A viscous fuel would lubricate and isolate joints better, but would be more easily lost through filter cleaning. The optimum kinematic viscosity at 40 °C is 2–6 mm²/s. This range of fuel viscosity will prevent high suction pressure (Jin *et al.* 2011).

Some characteristics of the fuel mixture components are listed in Table 3. Compared to standard biodiesel fuel (RME), RBE features additional positive characteristics, such as a low CFPP, which is even lower than that of the tested D.

Based on these findings, replacing RME with RBE in the fuel mixtures will likely improve the low-temperature performance and fluidity characteristics of the fuel, thus enabling better performance in colder climate and potentially eliminating the need for additives that improve low-temperature performance. RBE density and kinematic viscosity are slightly higher than those of RME, while the cetane number and the calorific value of RBE exceed those of RME. The values of cetane number for RBE and RME are higher than those observed for D, but calorific values are lower, it is likely that the use of pure RBE or the use of RBE in mixtures with D would increase fuel consumption, as is the case with RME–diesel mixtures.

Table 3. Main characteristics of fuel components

Fuel	CFPP (°C)	Density at 15 °C (kg/m ³)	Kinematic viscosity at 40 °C (mm ² /s)	Cetane number	Calorific value, (MJ/kg)
RME	–9	875	4.6	53.0	40.1
RBE	–16	888	5.6	59.7	40.6
D	–9	834	2.5	51.5	45.4

It should be noted that RME and RBE have a higher kinematic viscosity and density compared to D. This difference is typical for most biofuels because they originate from natural ‘fuels’ with large fatty acid ester carbon chain length, unsaturation levels and remnants of mono-, di- and triglycerides. Biodiesel fuel quality standards provide higher values of the above mentioned indicators compared to diesel. The fatty acid methyl esters density value at 15 °C ranges from 860 to 900 kg/m³, and the kinematic viscosity value at 40 °C ranges from 3.5 to 5 mm²/s for biodiesel. The RBE density and viscosity values are slightly higher (compared to RME) due to the higher molecular mass of alcohol employed for transesterification and thus the longer carbon chain of the esters is achieved.

Similar trends have been observed from data related to the physical and chemical characteristics of RBE- and RME–diesel mixtures. Data of Table 4 shows that mixtures containing RBE are characterised by slightly higher viscosity and density; however, they are compliant with the requirements of European Standards EN 590:2009+A1:2010 or EN 14214:2012, and their kinematic viscosity matches the optimal value of 2–5.0 mm²/s above.

Mixtures with higher levels of RBE are characterised by higher kinematic viscosity and density compared to mixtures with RME. With regard to the calorific value and cetane number, diesel mixtures containing RBE differ only slightly from mixtures containing RME; however, the calorific value of the mixtures is as much as 2% lower than that of D, which may contribute to higher fuel consumption.

Elemental composition of fuel is one characteristic that helps define and shape other characteristics. Conventional diesel that is used in internal combustion engines is composed primarily of carbon and hydrogen. During combustion, the elements react with oxygen, and the discharged heat is transferred to mechanical performance. Biodiesel fuel, contains higher oxygen content than D (in the case of RME, approximately 10.8% (mass), and in case of RBE, approximately 9.5%). Thus, in the case of biofuel,

Table 4. Physical and chemical characteristics of fuel mixtures

Parameter	Unit	EN 14214	EN 590	10% RBE	20% RBE	30% RBE	10% RME	20% RME	30% RME
Ester content	% (m/m)	min. 96.5	max. 5	10	20	30	10	20	30
CFPP	°C	Grade C, max. –5 °C	Grade C, max. –5 °C	–11	–12	–13	–9	–9	–9
Density at 15 °C	kg/m ³	860–900	820–845	839	843	846	837	845	854
Viscosity at 40 °C	mm ² /s	3.5–5.0	2.0–4.5	2.7	3.0	3.3	2.8	3.2	3.8
Cetane number		min. 51	min. 51	52.5	53.4	54.1	51.6	51.8	52.0
Calorific value	MJ/kg			45.0	44.5	44.0	44.8	44.3	44.0

Table 5. Elemental composition of the fuel

Fuel	C (%)	H (%)	O (%)
Conventional diesel	86.9	12.7	0.4
RME	77.1	12.1	10.8
RBE	78.1	12.4	9.5
10% RBE	86.1	12.6	1.3
20% RBE	85.2	12.6	2.2
30% RBE	84.3	12.5	3.1
10% RME	86.1	12.5	1.4
20% RME	84.9	12.6	2.5
30% RME	84.1	12.4	3.5

combustion processes are characterised by a higher excess air coefficient and thus a smaller amount of incomplete combustion products in the exhaust gases. Biodiesel fuel contains less carbon than D, which is attributed to the poorer thermal behaviour to biodiesel. It should be noted that biofuel and fossil fuel differ from each other in their fractional composition: the fatty acid esters in biodiesels are smoother (which can be proven by narrow interval of fatty acid distillation temperatures; in the case of RME, it is $299 \div 346$ °C), while D is fractionated by distillation at $177 \div 346$ °C.

The elemental composition of the tested fuel mixtures is presented in Table 5.

The results demonstrate that RME or RBE additives increase the oxygen content in the fuel, and the increase is more significant in the case RME than in the case of RBE. For a 30% addition of RME or RBE to D, the oxygen content is as much as $7.8 \div 8.8$ times higher (compared to diesel alone). The hydrogen levels in fuel mixtures containing $10 \div 30\%$ biofuel are comparable, and carbon levels decrease in proportion to the ester content in mixture.

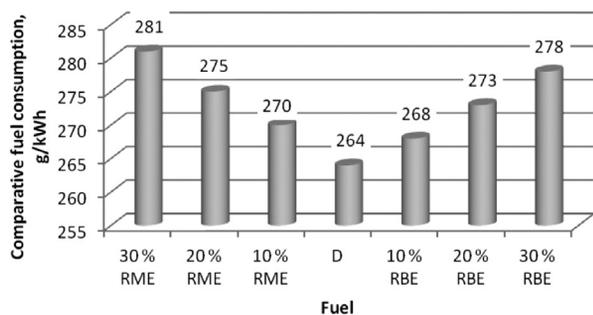


Fig. 1. Comparative fuel consumption at 2500 min^{-1} engine speed and 100 Nm engine load

2.2. Parameters of the diesel engine running on fuel mixtures

The results of investigation are presented in Figs 1–7.

In the comparison to the fuel consumption of biofuel versus pure diesel, there is a tendency for both RME- and RBE–diesel mixtures' growth up to 6% and up to 5%, respectively (Fig. 1). This tendency can be explained by the poor lower heating value of both esters (the difference between RME and diesel was 12%, and the difference between RBE and diesel was 11%). The difference in bottom thermal behaviour and thus fuel consumption for RME–diesel mixture, RBE–diesel mixtures and pure diesel is determined by their production technology: the use of alcohols with different cetane numbers for transesterification (Matijošius, Sokolovskij 2009). The physical and chemical characteristics of butanol are closer to those of D than methanol, and these characteristics contribute to its poorer bottom thermal behaviour and directly affect the comparative fuel consumption. Such findings are consistent with other biodiesel studies demonstrating that an increase in the bio-component concentration in diesel mixtures also increases comparative fuel consumption (Tsolakis *et al.* 2007; Valentino *et al.* 2011).

The engine was tested at 2000 and 2500 min^{-1} , at 20 , 40 , 60 , 80 and 100 Nm load, but we choose only one more characteristic mode, because of a huge quantity of data.

A higher cetane number compared to D (for D value is 51.5 , for RME value is 53.0 and for RBE value is 59.7) causes a shorter combustibility delay; however, a higher ester boiling temperature compared to diesel reduces the amount of prepared combustible mixture and thus causes lower thermal discharge in combustion phase 1. The lower thermal discharge then intensifies the combustion process in combustion phase 2 by improving the efficiency values (up to 3% in both RME and RBE cases) (Fig. 2) and reduces the amount of incomplete combustion product (CO,

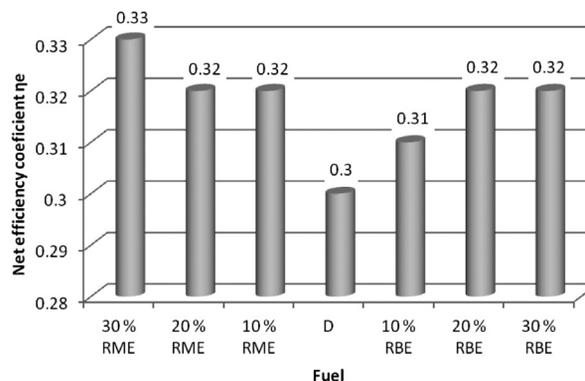


Fig. 2. Net efficiency coefficient at 2500 min^{-1} engine speed and 100 Nm engine load

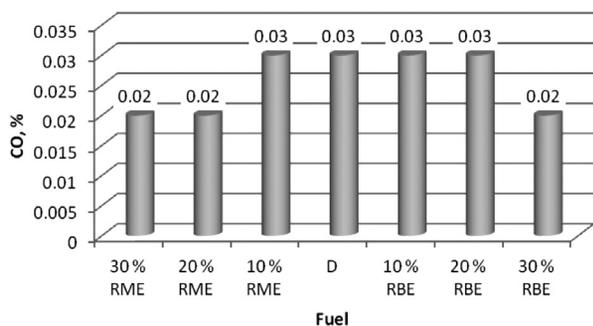


Fig. 3. CO gas emission at 2500 min⁻¹ engine speed and 100 Nm engine load

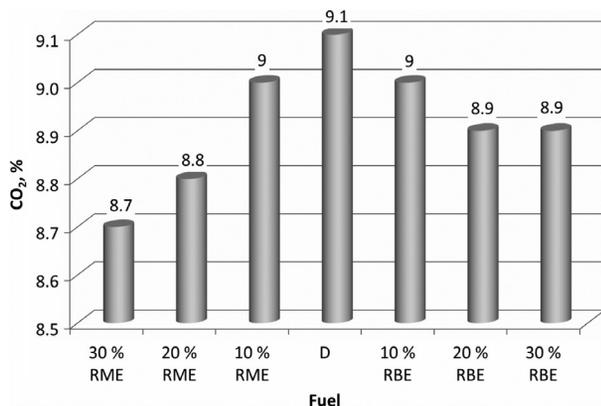


Fig. 4. CO₂ gas emission at 2500 min⁻¹ engine speed and 100 Nm engine load

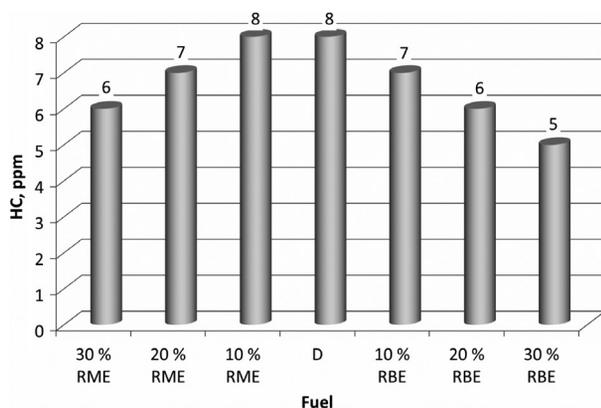


Fig. 5. HC gas emission at 2500 min⁻¹ engine speed and 100 Nm engine load

Fig. 3, HC, Fig. 5 and smoke, Fig. 7) concentrations in the exhaust gas.

Carbon monoxide (CO) emissions (Fig. 3) decreased in parallel to increasing concentrations of both esters in mixtures (RME and RBE, respectively) by up to 33%. This decrease does not contradict what is expected based on research in the literature (Corkwell *et al.* 2003; Rakopoulos *et al.* 2008, 2010).

Carbon dioxide (CO₂) emission decreases of 3% (RME) and 1% (RBE) (Fig. 4) are related to lower

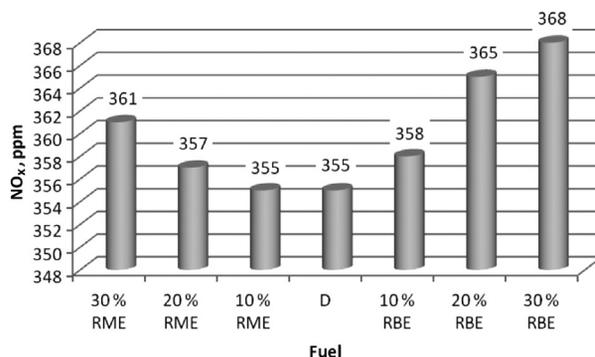


Fig. 6. NO_x gas emission at 2500 min⁻¹ engine speed and 100 Nm engine load

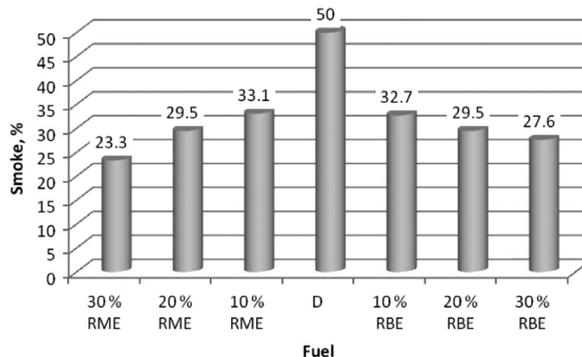


Fig. 7. Smoke emission at 2500 min⁻¹ engine speed and 100 Nm engine load

levels of carbon atoms in both esters compared to diesel alone.

With regard to the HC concentration (Fig. 5), a decrease in emissions has been observed with both RME (up to 25%) and RBE (up to 38%) mixtures; this decrease can be explained by a more intense combustion process in the engine cylinder when the working mixture disperses in the combustion chamber more evenly.

General emissions of NO_x have increased by up to 2% and 4% in conjunction with the increase in bio-component levels in RME and RBE compounds. This increase is related to the higher maximum pressure, and thus the higher temperature, that develops in the cylinder, causing high-temperature NO_x to form. These findings support the findings of Nagase and Funatsu (1990), which demonstrated that a major proportion of NO_x emissions in the cylinder are produced before the maximum pressure is reached.

Smokiness decreased using RME and RBE by up to 53% and 49%, respectively, due to the increased oxygen content in the fuel mixtures (Fig. 7); this finding relates to improvements in the combustion process and supports the idea that greater oxygen content in the compound results in the generation of less smoke.

Conclusions

The physical and chemical characteristics of RBE differ little from those of RME.

Comparative fuel consumption increased due to the differing thermal behaviour of the bio-component additives compared to D: in the case of 30% RME, the increase was 6%, and in the case of 30% RBE, it was 5%.

The addition of up to 30% bio-components to diesel improves the energy characteristics of the fuel, as illustrated by the higher effective efficiency (3% in cases of RME and RBE additives) compared to pure diesel.

Increasing the concentration of bio-components up to 30% yields a complex improvement in the environmental impacts of RME- and RBE-diesel mixtures: CO gas emissions decrease to 33% using additions of RME or RBE; CO₂ emissions decrease to 3% in the case of RME and to 1% in the case of RBE; HC emissions decrease to 25% in the case of RME and to 38% in the case of RBE; smokiness decreases to 53% for RME–diesel mixtures and to 49% for RBE–diesel mixtures. However, increases in NO_x emissions were observed with both RME–biodiesel mixtures (2%) and RBE–biodiesel mixtures.

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