Improvement of exhaust emissions in a diesel engine with the addition of an oxygenated additive to diesel-biodiesel blends

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Emissions control in internal combustion engines is the big challenge faced by engine manufacturers. Modern internal combustion engines exploit various systems to reduce exhaust emissions. However, the existing emission control systems will fall short of meeting stringent future emission regulations. This study attempts to reduce the exhaust emissions of a diesel engine fuelled with diesel-biodiesel blends by utilising ethyl acetate as a renewable oxygenated fuel additive. In this context, initially, ethyl acetate is mixed with biodiesel-diesel blends by 5% and 10% volume to obtain test fuels. Then, their fuel properties are measured by applying test methods proposed in the standards. Subsequently, engine experiments are conducted on a single-cylinder four-stroke diesel engine operated on distinct test conditions. The findings indicate that the inclusion of ethyl acetate in the diesel-biodiesel blends improves the fuel quality and markedly decreases emissions. A substantial reduction is achieved in NO_x, soot, and CO emissions up to 50%, 70%, and 71%, respectively, with a slight increase in fuel consumption in the case of adding ethyl acetate. More importantly, the addition of ethyl acetate enhances the NO_x-smoke trade-off and NO_x-BSFC trade-off characteristic of a diesel engine without loss of thermal efficiency. From this research, it can be inferred that ethyl acetate can potentially reduce exhaust emissions of the existing diesel engines fuelled with diesel-biodiesel blends.

Keywords: exhaust emissions, ethyl acetate, NO_x-PM tradeoff, oxygenated fuel

INTRODUCTION

Global warming is one of the biggest challenges of this century since it affects everything from climate to the economy. It is driven by mainly industrial facilities and transportation generating greenhouse gas emissions caused by burning fossil fuels [1]. For this reason, it is necessary to ensure abandoning fossil fuels that cause climate change and a progressive transition to renewable and clean energy. Transportation that includes road transport, aviation, shipping, rail, and pipeline is mainly powered by internal combustion engines (ICEs) and is responsible for approximately 16.2% of global greenhouse gas (GHG) emissions [2, 3]. ICEs are also used in other areas such as agriculture, construction, heat, and electricity generation. This indicates that ICEs are a great contributor to GHG and air pollution resulting from the combustion of fossil fuels. Fossil fuels are still the dominant energy source of global transportation; however, some countries are making progress in increasing the share of renewable fuels in the sector since using renewable fuels rather than fossil fuels in transportation is crucial to reduce the environmental impacts of exhaust gases [4]. Using renewable fuels not only reduces GHG and other harmful emissions but also reduces the dependence on imported oil, and thus it would ensure the countries' energy security. In this regard, governments enforce policies to increase the share of renewable fuels used in transport. For example, the European Union has set targets through Renewable Energy Directives [5, 6], whereby 10% and 14% of all energy used in transport is produced from renewable sources by 2020 and 2030, respectively. Similarly, in Turkey, biodiesel produced from local sources or waste oils can be blended with petroleum diesel fuel by up to 7% by volume [7]. Additionally, ethanol and methanol can be blended with gasoline by up to 5% and 3% volume, respectively [8]. Renewable alternative fuels are produced from biomass or waste and, currently, biodiesel, hydrotreated vegetable oil, and alcohols receive maximum attention and are widely used [9].

The use of oxygen-containing fuels acts as an oxidiser in combustion, offering a significant reduction in exhaust emissions without a substantial deterioration in engine performance compared to non-oxygenated fuels [10]. In addition, oxygenated fuels can be produced from a variety of renewable sources, yielding a decrease in GHG emissions and other pollutants [11]. Besides, the utilisation of oxygen-containing fuels for emission-reduction purposes is a more effective and less costly method than other emission control systems due to it not requiring any structural modification in the existing engine [12]. Thus, the application of oxygenated fuels in internal combustion engines is highly advantageous.

Many studies have researched the use of various oxygenated fuels with diesel or diesel-biodiesel blends and have concluded a promising improvement in the exhaust emissions of diesel engines. For instance, Agarwal et al. [13] observed lower particulate matter and NO_x emissions for diethyl ether-diesel blends. Singh et al. [14] found that butanol could decrease NO_x emission by 25% thanks to its cooling impact; they suggested that butanol can be effectively used with a diesel-biodiesel blend in diesel engines. Devarajan et al. [15] conducted engine experiments using n-decanol and di-tetra-butyl-peroxide as oxygenated additives to diesel-biodiesel blends. They concluded that the addition of these oxygenated additives to diesel-biodiesel blends by 10% volume could improve exhaust emissions and engine performance. Rahiman et al. [16] reported that di-butyl ether and ethanol provided substantial improvements in engine performance and exhaust emissions compared to diesel fuel. Sajin et al. [17] observed a decrease in CO, HC, NO_x, and smoke emission by the addition of dimethyl ether to biodiesel at 10% and 20% volume fraction. Yeşilyurt and Aydın [18] experimentally investigated the effect of diethyl ether (DEE) as an oxygenated fuel additive to diesel-biodiesel blend on combustion, performanceand emission characteristics of a diesel engine. The authors reported that the addition of the DEE by volume of 2.5%, 5%, 7.5%, and 10% in the biodiesel-diesel blend adversely affected the engine performance but it led to improving the exhaust emission. DEE provides a decrease of up to 8.84%, 4.12%, and 12.89% in NO_x, smoke, and HC emission, on average, compared to diesel fuel. Moreover, CO emission is mitigated by 40.09% at a low concentration of DEE, along with a reduction in CO₂ emission under high engine loads with DEE usage. Consequently, it is concluded that DEE can be blended up to 10% volume, which is recommended as a promising way to use biodiesel/diesel blend efficiently in diesel engines. Mehta et al. [19] reported that the addition of ethylene glycol monoacetate as an oxygenated fuel to the diesel-biodiesel blend improved the emissions characteristics of the engine. The ethylene glycol monoacetate in a volumetric fraction of 1%, 3%, 5%, and 7% were added to a diesel-cottonseed biodiesel blend, and the results showed that the higher oxygen concentration in the fuel mixture provided complete combustion and resulted in a substantial decrease in CO, HC, smoke, and NO_x emission. Zhang et al. [20]

conducted a simulation study on the emission characteristics of a diesel engine fuelled with diesel/methanol and ethanol-blended fuel using a three-dimensional CFD model. The simulation result showed that the diesel/methanol/n-butanol blends could reduce CO, HC, NO_x , and soot emissions. It was found that the optimal blending ratio of diesel, methanol and n-butanol is 70%, 20%, and 10%, respectively. Therefore, it was concluded that the diesel/methanol/n-butanol blended fuel could improve the combustion and emission characteristics of the engine.

Ethyl acetate, also known as ethyl ethanoate, is an organic compound with the formula of $C_4H_8O_2$. It is a colourless liquid, and it has a characteristic odour. Ethyl acetate is produced from the esterification reaction of acetic acid with ethanol as a conventional production method [21], and it has a wide range of applications, mostly as solvents in many industrial processes [22]. Besides, ethyl acetate can also be used as a renewable oxygenated fuel due to its favourable fuel properties such as high oxygen content, low viscosity, low boiling point temperature, low toxicity, and its renewable and biodegradable nature [22-24]. However, in the scientific studies found in the available literature, ethyl acetate is mostly used as a cosolvent for diesel-alcohol blends [25] or gasoline-alcohol blends [26] and as a reactant for biodiesel production [27]. The use of ethyl acetate as an oxygenated fuel additive for the diesel-biodiesel blend is less explored. Therefore, in this research, ethyl acetate is used as an oxygenated fuel additive to diesel-biodiesel blends for reducing exhaust emissions.

This study aims to investigate the effect of ethyl acetate on reducing exhaust emissions of a direct fuel injection diesel engine fuelled with diesel-biodiesel blends. For that, ethyl acetate as an oxygenated fuel is added to a diesel-biodiesel blend by 5% and 10% volume. The blending ratio for ethyl acetate is limited to 10% to avoid low cetane number and poor fuel lubricity as well as high fuel cost. Besides, thanks to selected blending ratios, it does not require any structural modification in the engine. Once measuring fuel properties by following related standards, engine tests are performed on a single-cylinder research diesel engine under various operating conditions. Finally, the obtained results are compared with reference fuel under the same operating conditions. The results of this research could offer significant findings regarding the application of ethyl acetate as a fuel additive to diesel-biodiesel blends for diminishing exhaust emissions.

MATERIALS AND METHODS

Conventional diesel, biodiesel, and ethyl acetate were the base fuels for the preparation of the test fuels. Diesel fuel satisfying EN 590 standard was purchased from a fuel station (OPET, Samsun, Turkey). Biodiesel that meets the requirements of the EN 14214 standard was obtained from a local licensed biodiesel producer (DB Tarımsal Enerji A.Ş., İzmir, Turkey). Ethyl acetate with a purity of 99.5% was provided by a local chemical company (TEKKİM, İstanbul, Turkey). In this study, three kinds of fuels - a diesel-biodiesel blend (B20), and two ethyl acetate-containing fuels - were prepared and used. A reference fuel designated as B20 was exploited to obtain base data for comparison. The ethyl acetate-containing fuels were EA5 and EA10 prepared by adding 5% and 10% ethyl acetate to the diesel-biodiesel blend. Liquid measuring beakers were used to measure the volume of fuels. The measured fuels were poured directly into another cup with a volume of three litres, and fuel blends were mixed using a mechanic mixer at a stirring speed of 1200 rpm for ten minutes at room temperature. At the end of this process, a homogenous fuel mixture was obtained and subsequently subjected to engine tests. Table 1 shows the composition of test fuels. The biofuel fraction in the final blends was kept constant at 20% volume because engine manufacturers approved the use of biofuels up to 20% volume under warranty for their engines. Further, this biofuel blending ratio was preferred because it did not require modification on fuel injection system. Density, dynamic viscosity, lower heating value, cold filter plugging point temperature, and distillation temperatures of the test fuel were measured according to the corresponding test methods. Effects of fuels on exhaust emissions were investigated by performing engine tests at various brake power outputs (0.9, 1.7, 2.6, and 3.5 kW) at a constant engine speed of 1500 revolutions per minute (rpm). These test conditions correspond to low, medium, high, and full engine power output, respectively. To draw graphs, the brake power outputs at those test conditions were normalised as 0.25, 0.50, 0.75, and 1.00 in the order already mentioned. To put it more clearly, the normalised values are the ratios of the measured engine power at each test point (P_i) to the maximum engine power (P_{max}), for instance, the low load condition corresponds to 25% of maximum engine power and its normalised value is 0.25. The technical specification of the test engine is given in Table 2. The engine was coupled to a water-cooled eddy current dynamometer with an absorbing power capacity of 10 kW. The schematic diagram of the test system is presented in Fig. 1. TESTO 350-XL gas analyser was used for measuring exhaust emissions. Soot emission was gauged with the BOSCH BEA 070 smoke opacity device. The periodic maintenance and calibration of the exhaust emission devices were carried out by authorised services. Throughout the experimentation, exhaust emissions were measured without after-treatment and were presented as raw emissions in the next section. In each experiment, the test engine initially was run for 15 min to stabilize the engine by monitoring the engine coolant temperature.

Table 1. Fuels composition by volume

Once the engine was stable at desired engine power output, the measurements were recorded by a data acquisition system six times in one-minute intervals and the average data were calculated for consideration. The uncertainty analysis according to Holman's method [28] was carried out, and the results are given in Table 3.

RESULTS AND DISCUSSION

The measured physicochemical properties of the test fuels are listed in Table 4. The fuel properties affect and govern the combustion inside the cylinder, and hence they are significant for the analysis of combustion, performance, and emission characteristics of the engine. The addition of ethyl acetate to the diesel-biodiesel blend shows good miscibility and stability; no phase separation has been observed for six months. It can be seen that adding ethyl acetate substantially affects fuel properties and causes a decrease in kinematic viscosity, heating value, cold filter plugging point temperature, and distillation temperature. Kinematic viscosity is one of the most important fuel properties for diesel engines. Fuel with low viscosity enhances the atomisation process, resulting in better

Test Fuel	Diesel (%)	Biodiesel (%)	Ethyl acetate (%)
B20	80	20	0
EA5	80	15	5
EA10	80	10	10

	Specifications		
	Brand/model	Kirloskar/TV1 Multifuel, VCR	
	Туре	Four-stroke, natural aspirated	
	Cylinder volume	661 cc	
2	Bore/stroke	87.5 mm/110 mm	
	Compression ratio	17.5:1	
	Engine speed	1500 rpm	
	Maximum torque & power	21.8 Nm & 3.5 kW	
- Tiller	Fuel injection type	Direct-injection	
	Injection pressure & timing	200 bar & 23°C A bTDC	
Test engine	Cooling system	Closed-loop water-cooling	

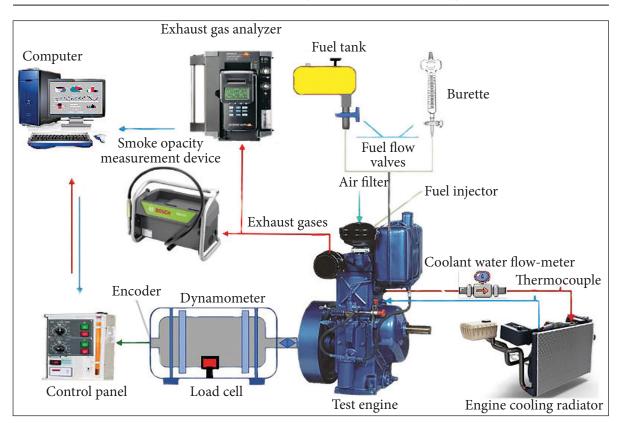


Fig. 1. Test setup arrangement

Parameters	Measurement Range	Resolution	Uncertainty		
Thermal efficiency	_	_	±0.96%		
Specific fuel consumption	_	_	±0.87%		
CO emission	0–10000 ppm	1 ppm	±10 ppm		
NO emission	0–3000 ppm	1 ppm	±5 ppm		
NO ₂ emission	0–500 ppm	1 ppm	±5 ppm		
Soot emission, k	0–9.99 m⁻¹	0.01 m ⁻¹	±1%		

Table 3. Uncertainties of the emission and performance parameters at rated engine power

fuel-air mixing and combustion, thereby improving engine performance and emissions. The addition of 5% and 10% ethyl acetate to the diesel-biodiesel blend decreases the kinematic viscosity by 25.7% and 36.9%, respectively. Ethyl acetate has a higher density (=906 kg/m³) and a lower boiling point temperature (=77.1°C) and molecular mass (=88.1 g/mol) than diesel and biodiesel fuel, thus it leads to an increase in density and a decrease in distillation temperature, as expected. The oxygen content in the ethyl acetate is 36.4% by mass, which improves the combustion of fuel but reduces the fuel heating value. Another significant improvement in fuel properties is the cold filter plugging point temperature, which is decreased down to -7° C by the addition of ethyl acetate, which indicates that ethyl acetate can also alleviate the trouble of cold flow properties of biodiesel.

The results of fuel characterisation have shown that the addition of ethyl acetate to diesel-biodiesel blends improves the basic fuel properties. Therefore, an improvement in emissions from the engine when ethyl acetate is incorporated into diesel-biodiesel blends can be expected. Exhaust emissions characteristics of the test engine are presented in graphs and discussed in the following paragraphs.

Properties	Test method	B20 Ref. [29]	EA5	EA10
Density @15°C (kg/m ³)	TS EN ISO 12185	844.5	847.7	850.8
Kinematic viscosity @40°C (mm ² /s)	DIN 53015	3.31	2.46	2.09
Lower heating value (kJ/kg)	ASTM D 240	41325	40388	39457
Cold filter plugging point (°C)	TS EN ISO 116	-5	-6	-7
Stoichiometric air-fuel ratio (kg/kg)	-	13.98	13.65	13.27
Distillation temperatures (°C)	TS EN ISO 3405			
	Initial boiling point	164.9	81.9	76.7
	10 vol.%	221.8	188.1	121.9
	50 vol.%	299.4	290.5	279.9
	90 vol.%	341.9	340.4	338.8
	95 vol.%	351.3	340.4	338.9
	Final boiling point	359.5	358.2	359.2
Recovered at 210°C, vol.%		6.8	12.9	19.4
Recovered at 250°C, vol.%, (limit: max. 60% vol.)		21.2	27.9	33.6
Recovered at 350°C, vol.%, (limit: min. 85% vol.)		94.5	94.9	95
95% vol. recovery temperature, °C, (limit: max. 360°C)		351.3	350.3	349.8

Table 4. Fuel properties

Incomplete combustion of the fuel results in unburned and partially burned products. Carbon monoxide (CO) occurs mainly due to a lack of oxygen in the combustion environment, poor fuel-air mixture, low combustion temperature, and insufficient combustion time [30]. The variation of CO emission for test fuels at selected engine power outputs is depicted in Fig. 2. As the engine power increases, the CO emission gradually decreases. This is due to the high in-cylinder temperature and turbulence intensity, which results in more complete combustion. CO emissions for ethyl acetate-blended fuels are significantly lower as compared to base fuel (B20), mainly because of the high oxygen fraction of ethyl acetate. Besides, the reduction in fuel viscosity results in high air-fuel mixing quality, leading to enhanced combustion, hence lower CO emission. It has been observed that adding 5% vol. ethyl acetate to the diesel-biodiesel blend can reduce CO emission by up to 71%. The EA5 and EA10 diminish the CO emission

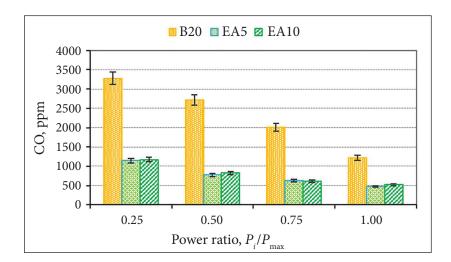


Fig. 2. Variation of CO emission for test fuels

by 67% and 66% on average compared to B20. Consequently, it is concluded that ethyl acetate might provide a great reduction in toxic CO emissions emitted from diesel engines.

The flame temperature, oxygen concentration, and combustion duration are the major factors in the formation of NO_x emissions in internal combustion engines [31]. An increase in the combustion temperature and oxygen fraction along with longer combustion period results in high NO_x emission; on the other hand, it may lead to better combustion. Besides, it is one of the most difficult emissions for controlling in practice since the precautions taken for reducing NO_x emissions generally result in a rise in other emissions or vice versa. Regarding NO_x emission given in Fig. 3, it is observed that NO_x emission increases with the rising engine power. That is caused by high flame temperature, which is deduced from the high exhaust temperature. According to the explanation given above, one can expect a rise in NO_x emissions when using ethyl acetate due to its high oxygen content. However, the outcomes of this study show that a substantial drop in NO_x emission is achieved with its utilisation. The ethyl acetate-blended fuels can reduce the NO_x emission by 50%. The average NO_x emissions for EA5 and EA10 are 36% and 40% lower than that of B20 fuel. It is because of a shorter ignition delay and lower calorific value of ethyl acetate than that of B20 fuel. It seems that these factors become more dominating than high oxygen content, bringing

about low combustion temperature and hence lower NO_x emission. The use of EA5 and EA10 lead to a shorter ignition delay period by 2°CA at full engine load compared to B20. A low ignition delay time causes a decrease in the amount of fuel burned in the premixed combustion stage and thus the NO_x formation rate slows. Besides, the low heating value of ethyl acetate causes to fall in the flame temperature, in turn, lower NO_x emission. An identical finding was observed by Razak et al. [12], who reported that adding alcohol to a diesel-biodiesel blend mitigated NO_x emission.

Soot/PM (particulate matter) emissions can be measured indirectly by a smoke opacity meter device, which gives information on the concentration of the particles existing in the exhaust gases. Soot/PM emissions are associated with environmental problems like air pollution, global warming, and damage to agricultural products [32]. The composition of particulate matter is very complex and mainly includes solid carbon particles, soluble and volatile organic substances, sulphate, and metals [33]. Therefore, soot emissions are toxic and carcinogenic. Emitted soot emissions harm human health, causing respiratory and nervous system diseases [34]. Figure 4 depicts the variation of smoke opacity for the test fuels at various engine power. Smoke opacity gradually augments as the engine power is increased irrespective of the fuel used for which the decrease in excess air coefficient is the main reason. The addition of ethyl acetate

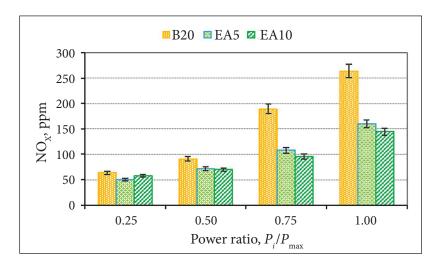


Fig. 3. Variation of NO_v emission for test fuels

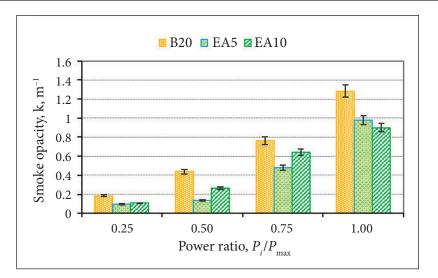


Fig. 4. Variation of smoke opacity for test fuels

to the diesel-biodiesel blend reduces the smoke opacity by 70%. This is mainly due to the high oxygen fraction in fuel composition. The simple chemical structure, low molecular mass, and low boiling point temperature of the ethyl acetate are the other reasons for the decrease in smoke opacity. Compared to B20, EA5 and EA10 decrease the average smoke opacity by 37% and 28%.

In conventional diesel engine combustion, there is a NO_x -soot (PM) trade-off and a NO_x -BSFC (brake-specific fuel consumption) tradeoff. Exhaust gas recirculation (EGR) and fuel injection timing retardation are the commonly used strategies for reducing NO_x emissions, however, in this case, an increase in soot emis-

sions and brake-specific fuel consumption occurs on account of diminished combustion temperature and oxygen concentration [35]. This is a substantial practical difficulty to meet increasingly strict emission regulations [36]. The trade-off between NO_x and smoke opacity for test fuels is depicted in Fig. 5. In this figure, as the curves approach the origin, the NO_vsmoke opacity trade-off improves. It is obvious from this figure that ethyl acetate-included fuels have a better NO_x-PM trade-off curve than that of B20 due to primarily high fuel oxygen. Therefore, ethyl acetate can simultaneously reduce NO_x and soot emissions. This result is similar to the one reported by Zhu et al. [36] and Agarwal et al. [13].

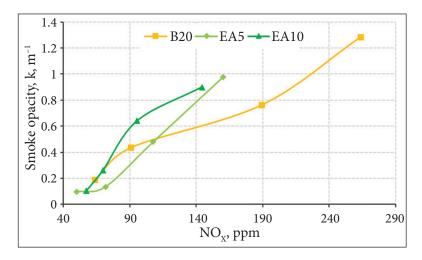


Fig. 5. NO_v-smoke opacity trade-off curve of test fuels

The NO_x-BSFC trade-off curve of test fuels is presented in Fig. 6. All fuels have somewhat better NO_x-BSFC trade-off at mid, high, and full engine power conditions than the low power condition because BSFC is the highest at low engine power output. It can be seen that the NO_x-BSFC trade-off curve for ethyl acetate-blended fuels is near to origin, which means that the change in both the NO_x emission and BSFC for EA5 and EA 10 is smaller than for B20. In other words, ethyl acetate-included fuels perform better in NO_x-BSFC trade-off performance than the reference fuel. Additionally, a higher ethyl acetate ratio can further reduce NO_x emission with little sacrifice in BSFC. This result is matched with that of Singh et al. [14], who noticed lower NO_x formation with a high butanol fraction.

Figure 7 presents the variation of BSFC and thermal efficiency for test fuels. BSFC increases with engine power, and an opposite trend occurs in thermal efficiency. This is basically due to the reduction in heat losses and an increase in combustion quality. As for the impact of test fuels on the engine performance characteristics, it has been found that ethyl acetate leads to an increase in BSFC due to its high density and

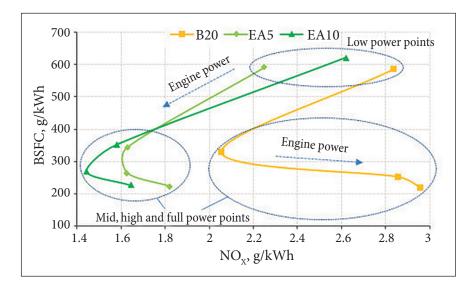


Fig. 6. NO_y-BSFC trade-off curve of test fuels

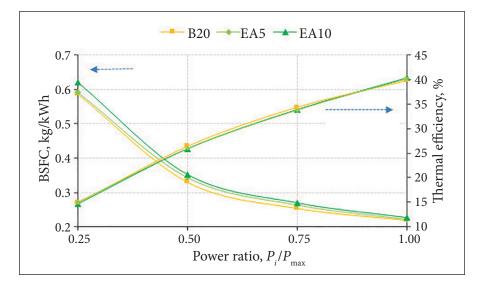


Fig. 7. Variation of BSFC and thermal efficiency for test fuels

low calorific value. On the other hand, no significant changes in thermal efficiency are observed. The reason for this can be due to lower kinematic viscosity and low boiling point temperature of ethyl acetate, resulting in an enhanced air-fuel mixture inside the cylinder, and subsequently, in enhanced combustion. The average BSFC for EA5 and EA10 is 2.3% and 5.8% higher than that for B20. Further, the differences in thermal efficiency are lower than its uncertainty value. Similar increases in brake-specific fuel consumption for oxygenated blended fuels were reported by Singh et al. [14], Ravi and Karthikeyanand [37], and Seelam et al. [38]. The main reason for that is attributed to the low calorific value of the oxygenated fuels. However, dissimilar findings were reported by Agarwal et al. [39] and Devarajan et al. [15]. They claimed that engine performance increased with the use of oxygenated fuel owing to efficient combustion. Moreover, some authors [40, 41] found that no significant variation in engine performance occurred with the use of oxygenated fuels.

CONCLUSIONS

In the present research, ethyl acetate, which is an oxygenated renewable fuel additive, is added to biodiesel-diesel blends by 5% and 10% volume to scrutinise its effect on reducing exhaust emissions. It was found that diesel-biodiesel blends containing ethyl acetate exhibited far superior fuel properties to reference fuel. Besides, ethyl acetate offers good miscibility with the diesel-biodiesel blend. Regarding exhaust emission characteristics, when ethyl acetate is added to diesel-biodiesel blends, a substantial reduction is detected in CO, NO_x, and soot emissions up to 71%, 50%, and 70%, respectively, against an increase in BSFC by 5.8%. Moreover, a remarkable enhancement of NOx-PM trade-off and NO_x-BSFC trade-off is achieved through ethyl acetate use with keeping the same thermal efficiency. During the experimentation, no engine functionality problems were encountered, and it was found that ethyl acetate was compatible with the diesel-biodiesel blend. Considering all findings, it can be concluded that ethyl acetate is a promising component for use in diesel engines owing to its significant potential to reduce exhaust emissions without any engine modification but with a bit of sacrifice in brake-specific fuel consumption. However, further investigations concerning the transient running conditions of the engine are necessary. Besides, investigations on optimising the engine settings or reprogramming of the engine control unit as well as modifications of mechanical parts of the engine to yield optimal performance with maintaining low emissions should be carried out.

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Abdülvahap Çakmak

DYZELINIO VARIKLIO IŠMETAMŲJŲ TERŠALŲ KIEKIO GERINIMAS Į DYZELINO IR BIODYZELINO MIŠINIUS PRIDEDANT DEGUONIMI PRISOTINTO PRIEDO