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The effect of a homogeneous combustion catalyst on exhaust emissions from a single cylinder diesel engine

Yu Ma, Mingming Zhu, Dongke Zhang *

Centre for Energy (M473), The University of Western Australia, 35 Stirling Highway, Crawley, WA 6009, Australia

HIGHLIGHTS

- ▶ Effect of a Fe-based homogeneous catalyst on diesel engine emissions was studied.
- ▶ The fuel properties were not altered by the addition of the catalyst.
- ▶ The catalyst led to 4% fuel saving under the tested conditions.
- ▶ The catalyst reduced engine emissions by up to 39% smoke, 21% CO and 13% UHC.

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ABSTRACT

This paper reports a series of experimental investigations into the effect of an Fe-based homogeneous combustion catalyst on the emission characteristics from a four-stroke single cylinder diesel engine. The catalyst contained ferrous picrate as the active ingredient in a composite organic solvent mixture which could be homogeneously dissolved into a commercial diesel fuel at ultra low dosage ratios. The engine tests were conducted at four different engine loads and at two steady speeds of 2800 rpm and 3200 rpm, respectively. Engine exhaust emissions of CO, unburnt hydrocarbons (UHCs) and NOx were measured using an AVL gas analyser and the particulate emissions were evaluated in terms of smoke opacity using a Bosch smoke meter. The results showed that, in addition to the benefit of improved fuel efficiency, the homogeneous combustion catalyst significantly reduced the emissions of particulate matter, CO and UHC from diesel engines. Compared with the reference diesel, up to 3.7% reduction in the brake specific fuel consumption was achieved when the diesel fuel was treated with the catalyst. The use of the catalyst also led to significant reductions in the particulate matter, CO and UHC emissions, with the maximum reductions being 39.5%, 21.1% and 13.1%, respectively. The NOx emissions, however, increased slightly, by ca. 6%, which was consistent with the improved combustion performance as reasonably expected.

1. Introduction

Compression ignition (CI) engines, also known as diesel engines, are widely used in transportation, heavy machinery and power generation due to their higher efficiencies, power outputs and durability compared with spark ignition petrol engines. However, the ever-increasing demand for diesel engines has also led to increased emissions of a range of pollutants with adverse effects on the environment and human health [1,2]. Diesel engine exhausts are typically severe, which contain, depending on the engine design and operational parameters, large amounts of particulate matter (PM) or smoke emissions and varying amounts of carbon monoxide (CO), unburned hydrocarbons (UHCs), nitrogen oxides (NOx), sulphur oxides (SOx) and carbon dioxide (CO_2) [3,4]. Recently, under the enforcement of increasingly stringent emission regulations, advanced technologies to control diesel engine exhaust emissions have become essential and received more and more research and development interests [5–9].

Based on previous work, the addition of metal-based combustion catalysts to diesel fuel has shown to be an effective approach to improving diesel combustion, reducing fuel consumption and more importantly, lowering engine emissions [10–12]. At the molecular level, these catalysts consist of a metallic component as the active ingredient and a composite organic solvent which dissolve in liquid hydrocarbon fuels such as diesel, thus also named as homogeneous combustion catalysts (HCCs). These metal-based HCCs include platinum (Pt), cerium (Ce), manganese (Mn), magnesium (Mg), iron (Fe), nickel (Ni), calcium (Ca) and copper (Cu) [13–18]. Valentine et al. [13] studied the effect of a bimetallic Pt/Ce

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* Corresponding author. Tel.: +61 8 6488 7600; fax: +61 8 6488 7235.
E-mail address: dongke.zhang@uwa.edu.au (D. Zhang).

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catalyst on the engine performance when fuelled with diesel dosed with 4–8 ppm of the active metals and achieved 10–25% reduction in PM and 10–30% reductions in UHC and CO emissions, together with 5–7% improvement in fuel economy. Guru et al. [18] reported the influence of Mn and Mg based catalysts on the performance of diesel engines at the metal to fuel dosage ratio of 8 μmol/L and 16 μmol/L and found that the two catalysts were able to achieve the maximum reduction ratios of 3.1% and 2.0% for fuel consumption, 16.4% and 13.4% for CO emission, 29.8% and 17.9% for PM emission, respectively. May and Hirs [19] conducted a series of engine tests by using an Fe/Mg bimetallic-organo combustion catalyst containing 50 ppm Fe and 10 ppm Mg, respectively, and observed significant reductions in fuel consumption due to more efficient combustion with the catalyst treated fuel.

Among these aforementioned metals, iron has also been shown to play an important role in emission reductions. Since Shayeson [20] in 1967 for the first time detected the smoke reduction in a jet engine by applying iron compounds, various laboratory studies have been carried out to explore the effects of iron in premixed [21] or diffusion flames [22], drop tube furnace [23], boilers [19] and diesel engines [24,25], in the forms of iron pentacarbonyl ($Fe(CO)_5$), ferrocene ($Fe(C_5H_5)_2$), iron naphthenate, and iron chloride ($FeCl_3$). The present contribution details a laboratory study of the diesel engine emission characteristics involving an Fe-based HCC, which consists of an organometallic compound, ferrous picrate, as the active ingredient. The catalyst can be directly mixed with commercial diesel fuel at an ultra low dosage ratio. Field trials and our previous laboratory studies [26–28] have shown significant benefits in improved fuel economy and engine performance. It was found [27,28] that the catalyst can reduce the combustion duration of the fuel in the engine and result in a faster rate of heat release and higher peak cylinder pressure, thus enhancing the fuel combustion efficiency and saving the fuel consumption rates. However, there is a lack of systematic evaluations of the efficacy of the catalyst in controlling the engine emissions.

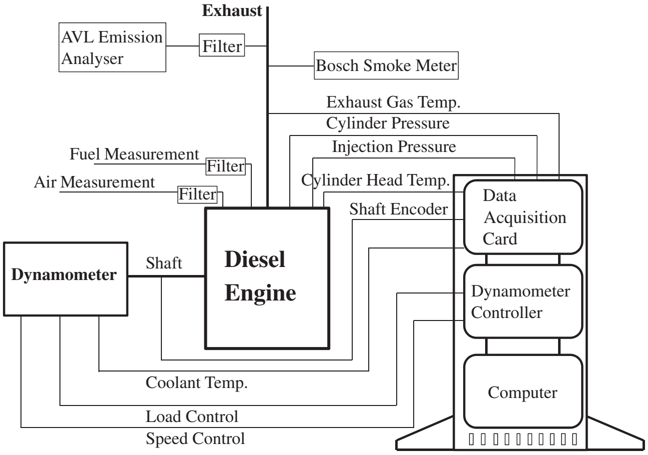

Fig. 1. Schematic diagram of the diesel engine system.

In the current study, diesel fuels, with or without the catalyst, were tested in a single cylinder diesel engine. Exhaust emissions including CO, UHC, NOx and PM were measured under various engine operating conditions.

2. Experimental

2.1. Test engine and instrumentation

The experiments were conducted on a single cylinder, naturally-aspirated, four-stroke, air-cooled, direct-injection YANMAR L48AE diesel engine (AET Ltd.). The engine had 70 mm bore, 55 mm stroke, 211 cm^3 displacement, compression ratio of 19.9:1 and was capable of delivering 3.5 kW rated brake power at the speed of 3600 rpm. A water-cooled, Zöllner A-100 electric dynamometer was directly coupled to the engine output shaft for providing various load conditions. Calibrated sensors/probes were installed to monitor the ambient and intake air temperatures, oil temperature, dynamometer coolant temperature and the exhaust temperature. A schematic layout of the diesel engine test system is shown in Fig. 1.

A 1L fuel tank sitting on a digital weighing scale (Acculab LT-3200) was connected to the fuel delivery line for measuring the instantaneous fuel consumption at a fixed time interval. For exhaust measurements, an AVL Digas 4000 emission analyser was used to analyse the concentrations of CO, UHC, and CO_2 by an infrared method and NOx and O_2 by an electrochemical method. PM emission measurement was achieved by monitoring the smoke opacity of the exhaust with a Bosch RTM 430 infrared opacimeter. SOx emission was not concerned in the present study since the compositions of the catalyst were not expected to introduce extra sulphur into diesel fuel. To ensure the high accuracy of each measurement, the instruments were carefully calibrated before each test. Table 1 shows the accuracies and ranges of the employed instruments for the emission measurements.

2.2. Fuels

A commercial diesel fuel was obtained from a local Caltex service station (Caltex Australia Ltd.) and used as the reference diesel in this study. Two homogeneous combustion catalysts, FTC and FPC, were provided by Fuel Technology Pty., Ltd. Both catalysts had similar compositions with different amounts of ferrous picrate as the active ingredient dissolved in a composite organic solvent made of short-chain alkyl benzene and its derivatives, which help improve the stability of the ferrous picrate–water–butanol–diesel mixture [22]. The Fe^{2+} contents in FTC and FPC were 180 mg/L and 560 mg/L, respectively. In the experimentation, the catalysts were respectively added into the reference diesel at two dosing ratios, namely, 1:3200 for FTC and 1:10 000 for FPC (by volume) due to the different concentrations of ferrous picrate in the FTC and FPC catalysts. For convenience, the reference diesel fuel was denoted as

Table 1
Specifications of the instruments used for the emission tests.

Instrumentation	Measurement	Measuring range	Accuracy	Measurement principle
AVL digas 4000	CO	0–10% by vol.	0.01%	Infrared measurement
	CO_2	0–20% by vol.	0.1%	
	UHC	0–20 000 ppm vol.	1 ppm	
	NOx	0–4000 ppm vol.	1 ppm	Electrochemical measurement
	O_2	0–4% by vol.	0.01%	
		4–22%	0.1%	
Bosch RTM 430 infrared opacimeter	Opacity	0–100%	0.1%	Optical LED measurement

Table 2
Key properties of the fuels used in the tests.

Parameters	RD	FTC-D (1:3200)	FPC-D (1:10000)	Analytical method
Viscosity (cSt at 40 °C)	2.03	1.98	2.02	ASTM D445
Density (kg/m^3 at 15 °C)	844.5	846.5	845.5	ASTM D1298
Flash point (°C)	80	78	81	ASTM D93
Pour point (°C)	−9	−8	−10	ASTM D97
Sulphur content (ppm)	<15	<15	<15	ASTM D1266
Distillation range (°C)	180–360	175–357	177–361	ASTM D86
Calculated cetane index (CCI)	48.7	48.5	48.8	ASTM D4737

“RD”, while the two catalyst-treated diesel fuels were abbreviated following the catalyst names and their dosage ratios, that is, “FTC-D (1:3200)” and “FPC-D (1:10000)”, respectively. The key physical properties of the three fuels were determined prior to the engine tests. As shown in Table 2, there were no noticeable changes to the diesel fuel properties after the addition of the catalysts.

2.3. Test procedure and data collection

The experiments were conducted with the three fuels under conditions of two engine speeds at 2800 rpm (low) and 3200 rpm (medium), respectively, and four engine loads at each engine speed, corresponding to 0.14 MPa, 0.21 MPa, 0.33 MPa and 0.42 MPa of the brake mean effective pressure (BMEP). Under each of the test conditions, the engine was allowed to operate at the specified speed and load for about 30 min until the characteristic temperatures (exhaust, lube oil and coolant) of the engine stabilized. Fuel consumption was measured by the weighing scale at 5 min intervals for each engine condition and automatically recorded by the computer. Exhaust emission measurements were taken directly from the exhaust pipe and the readings were recorded at 5 s intervals for 3 min continuous measurements. Each test was repeated three times and the results presented in this paper are the averaged values. For the tests involving a different fuel, the engine was purged with the new fuel for at least 30 min to eliminate the effect of the fuel in the previous test. The emission results were expressed in the units of grams of each pollutant per kg of the fuel burned and also grams of the pollutant per unit of the brake-specific power output following the procedures recommended by the EPA regulations, except for the smoke opacity. In order to reveal any statistically significant differences between the emission results from the reference diesel and the catalyst treated fuels, the completely randomised single-factor analysis of variance tests (ANOVA) [29] and the Turkey–Kramer tests [29] were applied. A 95% confidence level was used throughout the present study.

3. Results and discussion

3.1. Effect of FTC/FPC catalyst on fuel properties

The specifications of the fuels, with and without FTC/FPC catalysts added, are listed in Table 2. The test results showed no significant difference in the key fuel properties between the reference diesel and the two catalyst treated fuels, in terms of viscosity, density, flash point, distillation range and Cetane number index. This is easily understood because of the extremely low dosage ratios of the catalysts used. This follows that no modifications to the engine systems would be required in order to use the catalyst treated diesel fuels. Note that the invariant Cetane number also means that the ignition delay period would not be significantly influenced by the addition of the catalysts, as we already experimentally observed and reported elsewhere [28].

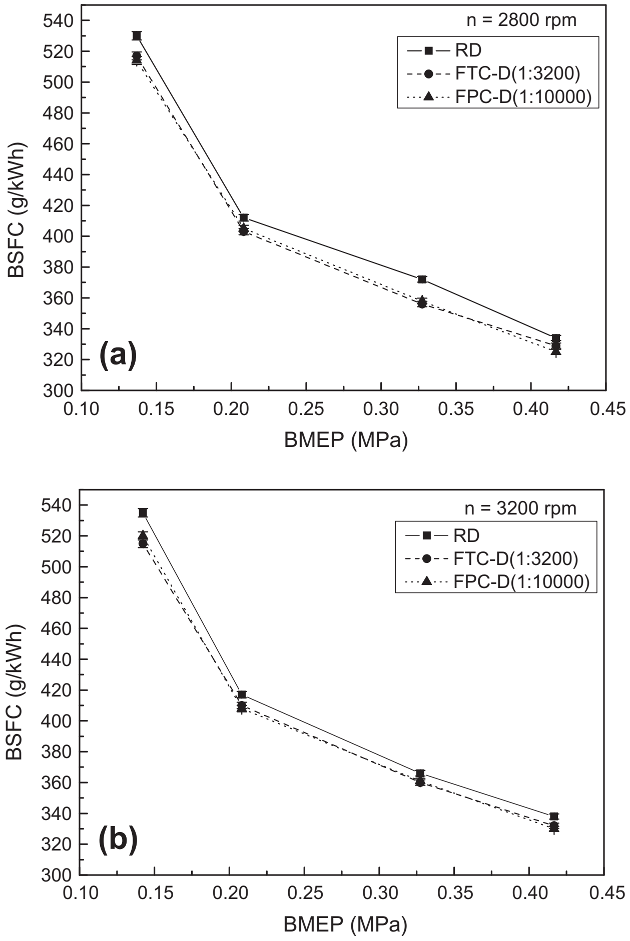

Fig. 2. Variation of brake specific fuel consumptions as a function of engine load for the three fuels at different engine speeds: (a) 2800 rpm and (b) 3200 rpm.

3.2. Effect of FTC/FPC catalyst on fuel consumption

For each engine test, the fuel consumption rate was measured as the fuel mass flow rate and based on which the brake specific fuel consumption (BSFC) was calculated. The BSFC value measures how efficiently the engine is producing work by using the fuel supplied [30]. A comparison of the BSFC for the reference diesel and the FTC/FPC catalyst treated fuels as a function of engine load at the two different engine speeds of 2800 rpm and 3200 rpm, respectively, are shown in Fig. 2. The BSFC was found to decrease with increasing engine load due to the improved fuel efficiency under the tested conditions, which means less fuel was burned to produce the same amount of work at higher loads. The BSFC values for the FTC/FPC catalyst treated fuels were found to be lower than that of the reference diesel under all engine test conditions. The maximum fuel saving of 3.7% was achieved for the FTC-D (1:3200) under the test condition of 3200 rpm and 0.14 MPa BMEP, while for the FPC-D (1:10000), the maximum fuel saving was 3.1% under the test condition of 2800 rpm and 0.14 MPa BMEP. These fuel saving values are comparable to our previous work on the effect of the FPC catalyst on the fuel efficiency [23,24] in which a maximum of 4.2% fuel saving was achieved under conditions of the engine load of 0.12 MPa and the FPC catalyst dosing ratio of 1:10000. The reduced fuel consumptions were due to the improved fuel combustion efficiencies of the engine operating with the catalyst treated fuels. This is explained by the authors' previous observations of higher peak flame temperature, increased cylinder pressure and faster heat release rate in the engine fuelled with diesel treated with the homogeneous combustion catalysts [27,28]. The mechanism of working of the catalyst in the diesel combustion process was experimentally studied with single fuel droplets in our previous work [31,32]. Ferrous picrate in the catalyst was found to decompose before the fuel vaporisation, allowing active iron atoms to be released to the combustion zone. Consequently an increased fuel burning rate and flame temperature were observed.

3.3. Effect of FTC/FPC catalyst on exhaust emissions

The results of emissions of PM, CO, UHC, and NOx are shown in Figs. 3–7, respectively. As mentioned before, SOx emissions were not studied in the present work as it is generally agreed that all sulphur contained in the diesel would be converted into SOx and sulphate in the engine combustion process [33] and the addition of the catalysts in this study did not change the sulphur content of the fuel.

3.3.1. Smoke emission

Smoke emission is a most visually noticeable pollutant from diesel engines which provides an indication of the particulate matters (PMs) levels [30]. Diesel PM is generally composed of agglomerated soot particles with volatile hydrocarbons adsorbed on the surfaces. As the main substance of the PM components, soot is considered to be responsible for the smoke opacity and its formation is due to incomplete combustion [30,34]. It was observed in the present work that the smoke opacity increased with increasing engine load, regardless if a catalyst was added in the fuel, as shown in Fig. 3. At high engine loads, more fuel was supplied at each engine cycle to engine of a fixed volume (displacement) of space for the combustion to take place. This means the residence time for fuel combustion is shortened, compounded with imperfect mixing of the fuel and combustion air, therefore, an increase in PM emission is expected at increased engine loads. Most interestingly, however, it was observed that the PM emission was dramatically suppressed by the addition of FTC/FPC catalysts to the diesel under all test conditions. The lowest smoke level was observed at 3200 rpm and 0.21 MPa BMEP for FTC-D (1:3200) and at 2800 rpm and 0.14 MPa BMEP for FPC-D (1:10000), with the maximum reduction ratios of 38.8% and 39.5%, respectively. The mean reductions in the PM emissions among all test conditions were 26.5% for FTC-D (1:3200) and 25.2% for FPC-D (1:10000), respectively.

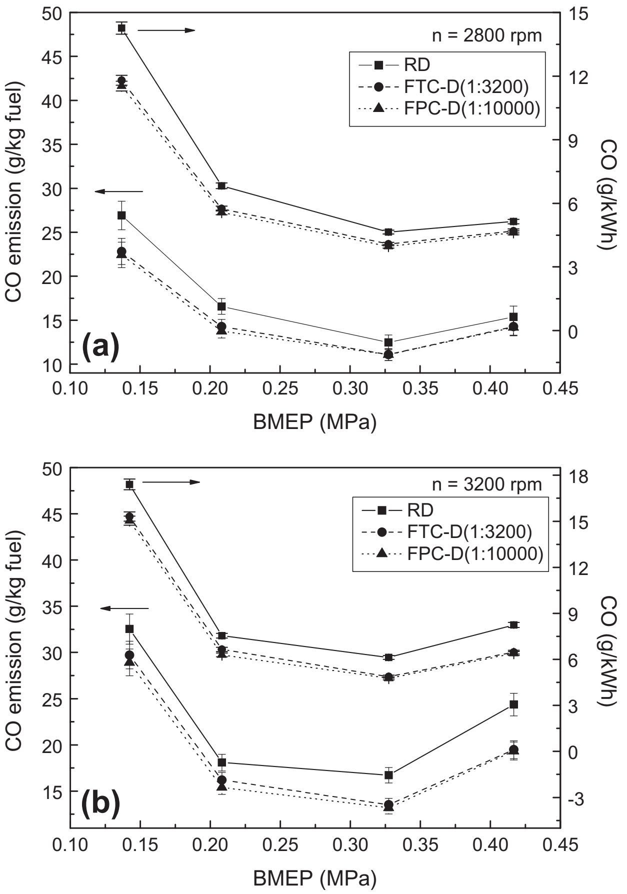

Fig. 4. Variation of CO emissions as a function of engine load for the three fuels at different engine speeds: (a) 2800 rpm and (b) 3200 rpm.

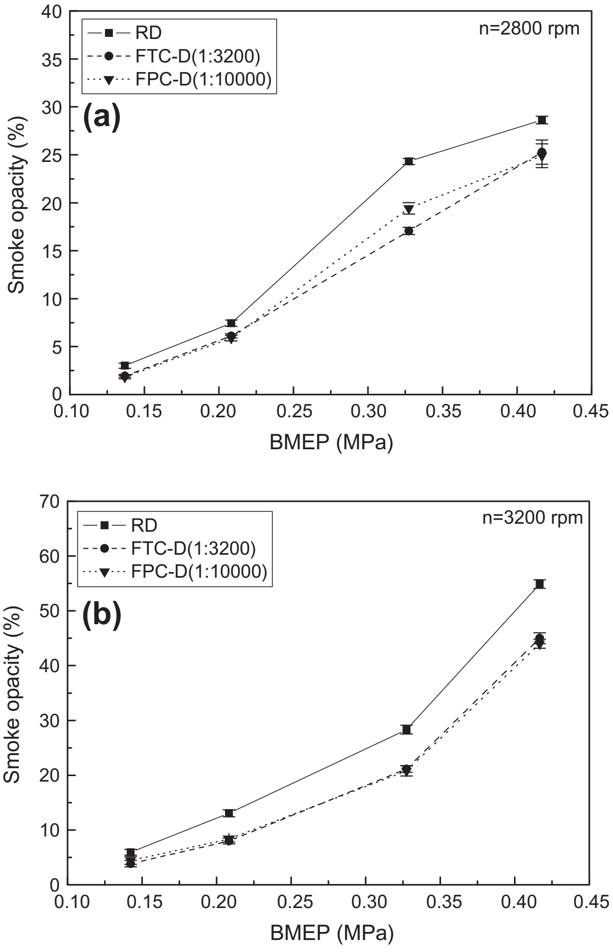

Fig. 3. Variation of smoke emissions as a function of engine load for the three fuels at different engine speeds: (a) 2800 rpm and (b) 3200 rpm.

The significantly reduced PM emissions are attributed to the enhanced combustion efficiency by the application of the catalysts. The effectiveness of iron in promoting soot oxidation and suppression has been reported in the literature [22,25,35–37]. Zhang and Megaridis [37] studied the mechanism of iron interfering with soot formation using ferrocene in a laminar non-premixed ethylene flame and proposed that the ferrocene decomposed early to form a fine aerosol for the subsequent carbon deposition. However, Hahn and co-workers [22] conducted experiments on isooctane diffusion flames with iron pentacarbonyl addition and found that

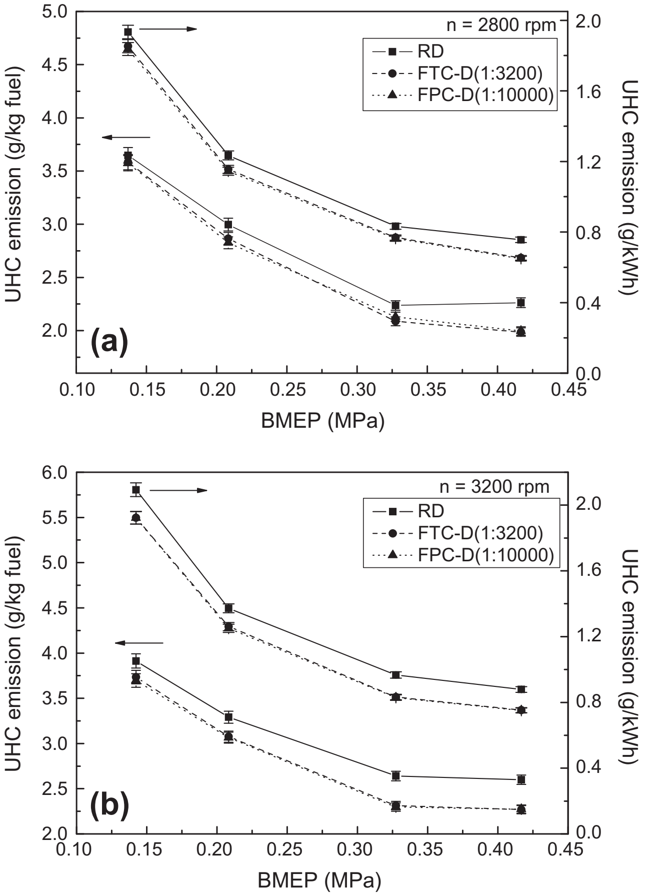

Fig. 5. Variation of unburned hydrocarbon (UHC) emissions as a function of engine load for the three fuels at different engine speeds: (a) 2800 rpm and (b) 3200 rpm.

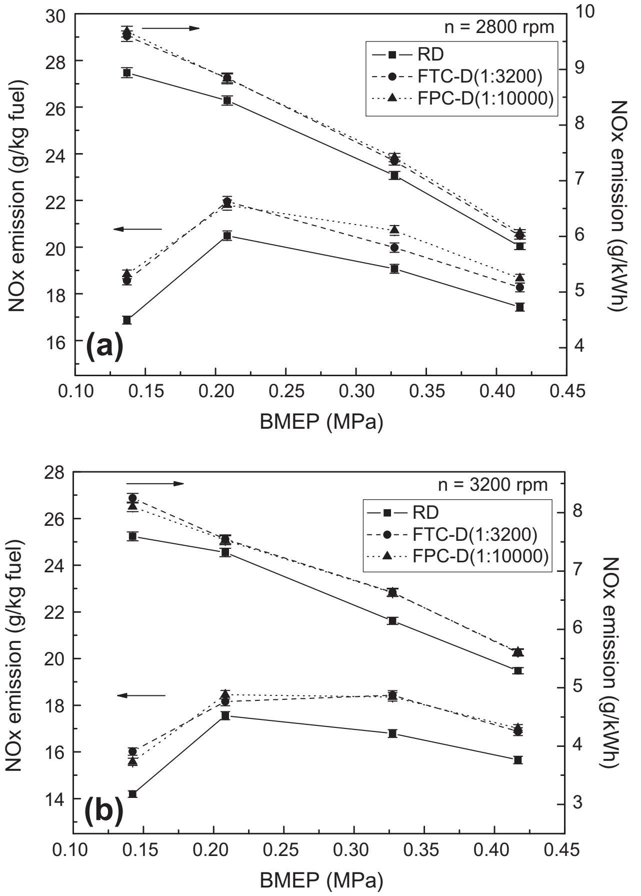

Fig. 6. Variation of NOx emissions as a function of engine load for the three fuels at different engine speeds: (a) 2800 rpm and (b) 3200 rpm.

rather than affecting soot particle inception and growth, iron was more effective in enhancing the soot oxidation during burnout and yielded a two-thirds reduction in the overall soot emissions. May and Hirs [19] reported an organo-metallic iron-based combustion catalyst for PM reductions in turbines combustion. It is believed that iron ions catalyses the oxidation of soot formed during diesel combustion resulting in a reduced PM emission [25,35–37].

3.3.2. *CO and UHC emissions*

CO and unburned hydrocarbons (UHCs) emissions are consequences of incomplete combustion of the fuel in diesel engines, especially when oxygen in local combustion zone is insufficient and/or the combustion temperature is low [30]. Generally, CO and UHC concentrations in the engine exhaust can be used as important parameters for determining the engine combustion inefficiency. Figs. 4 and 5 illustrate CO and UHC emissions, normalised as per kg of each fuel burned and brake power (kW), respectively. The two sets of curves show the same trend in the variations of the CO and UHC emissions versus engine load. It was observed that for all three fuels tested, the CO and UHC emissions tended to decrease with increasing the engine loads (except at the highest load – see below). This is due to the improved brake specific fuel efficiency and increased combustion temperature at high engine loads. However, the CO and UHC emissions increased again when the engine operated at the highest loads of 0.42 MPa BMEP. This is explained as that, under this highest load condition, in order to meet the high output power, a large amount of fuel was injected to the engine at each cycle which resulted in poor air–fuel mix and therefore an increase in the CO and UHC emissions.

The FTC and FPC catalysts are effective in reducing the incomplete combustion products of CO and UHC. Figs. 4 and 5 show that the CO and UHC emissions from the catalyst treated fuels were lower than those from the reference diesel under all test conditions. The maximum CO reductions observed were 20.1% for FTC-D (1:3200) when the engine operated at 3200 rpm and 0.42 MPa BMEP, and 21.1% for FPC-D (1:10000) at 3200 rpm and 0.33 MPa BMEP. The average CO reductions among all tested engine conditions were 13.2% for FTC-D (1:3200) and 15.1% for FPC-D (1:10000), respectively. The maximum UHC reductions achieved were 12.7% for FTC-D (1:3200) and 13.1% for FPC-D (1:10000), both at 3200 rpm, 0.42 MPa BMEP. The average UHC reductions among all testes were 7.7% and 7.8% for the two catalyst treated fuels. This is consistent with the observations of improved fuel efficiency and reduced PM emissions due to the addition of the FTC and FPC catalysts.

3.3.3. *NOx emission*

The oxides of nitrogen (NOx) are one of the critical emissions from combustion engines. NOx emitted from engines includes nitric oxide (NO) as the predominant compound and nitric dioxide (NO_2) in small quantities. The formation of NOx from diesel engines is due to the oxidation of the atmospheric nitrogen and is largely dependent on the peak flame temperature, oxygen concentration and residence time [30]. The influence of the FTC and FPC catalysts on NOx emissions at different engine loads are illustrated in Fig. 6. The NOx emission was found to be slightly increased for the catalyst treated fuels compared to the reference diesel under all test conditions. The maximum NOx levels were ob-

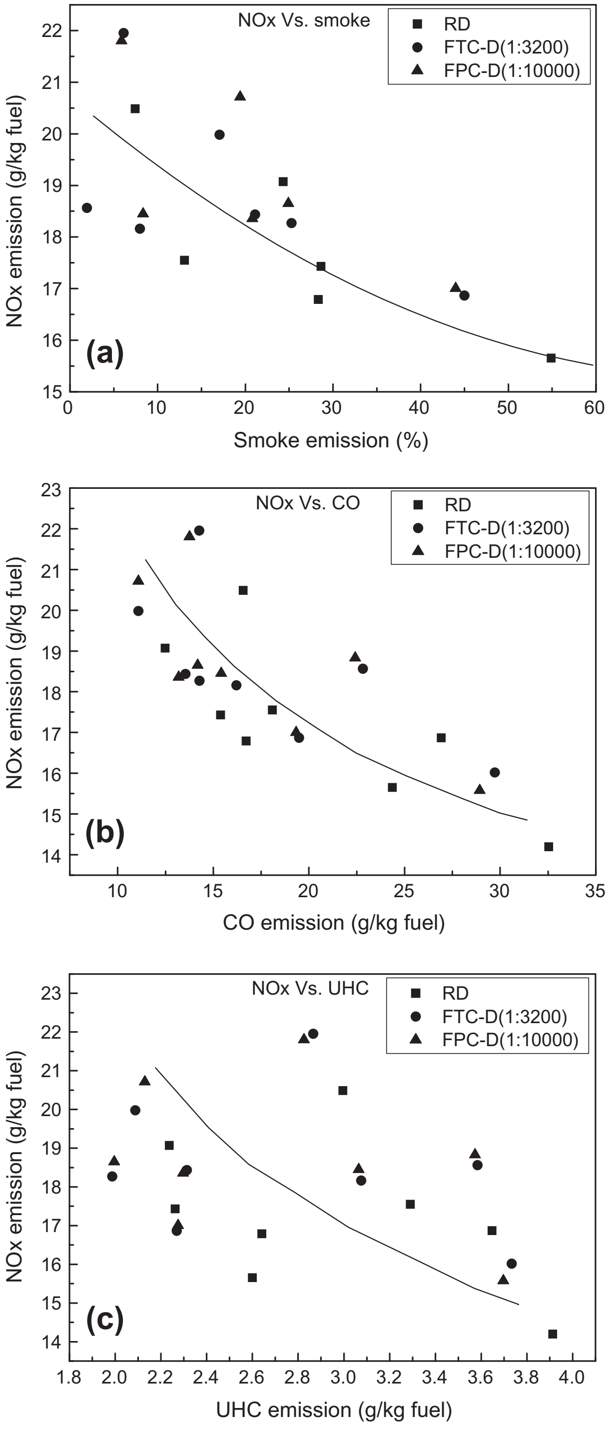

Fig. 7. The correlation between NOx and (a) smoke, (b) CO, and (c) UHC emissions, respectively.

served to be 21.9 g/kg fuel for FTC-D (1:3200) and 21.8 g/kg fuel for FPC-D (1:10000), compared to 20.5 g/kg fuel for the reference diesel at 2800 rpm and 0.21 MPa BMEP. These are 6.8% and 6.3% increases in the NOx due to the addition of the FTC and FPC catalysts, respectively, relative to the reference diesel. Increased NOx has also been reported by other researchers when applying iron-based catalyst in the form of $FeCl_3$ [24] and Mn/Mg metallic catalysts [18]. It may be explained that due to the presence of metallic catalysts, the fuel combustion is improved, resulting in a high flame temperature and cylinder pressure, which in turn result in more N_2 in the combustion air being oxidised to NOx during the combustion process [18,24,38]. In the authors' previous work [28], it was confirmed that higher combustion rates and flame temperatures as well as high engine cylinder pressures and heat release rates and, therefore, high combustion efficiencies, were achieved with the catalyst treated fuels.

3.3.4. Correlation of NOx versus smoke, CO and UHC emissions

It has been repeatedly observed that in a given combustion system, including diesel engines, increased NOx emissions are accompanied with reduced PM, CO and UHC emissions [30,39]. This is because the formation and emissions of these pollutants are all highly dependent on the combustion efficiency, where high combustion efficiency generates low PM, CO and UHC while also leads to high flame temperature and thus high NOx formation, and *vice versa* [40]. The present experimental results agree well with the literature reports and this well established mechanism [30,39,40]. Fig. 7 presents the correlation between the NOx emissions and the PM, CO and UHC emissions, respectively, obtained from the current engine tests. The emission results from both the reference diesel and the FTC and FPC catalyst treated fuels followed a similar trend, that is, when the engine operated more efficiently with less PM, CO and UHC emissions, more NOx was produced due to the increased combustion efficiency and thus higher flame temperatures. Adding the homogeneous combustion catalysts did not break the trade-off between NOx and the incomplete combustion emissions. However, the benefits of the use of the homogeneous combustion catalysts in improving the combustion efficiency and therefore, reduced fuel consumption and incomplete combustion pollutants for a given work done by the engine are clearly evident [18,24].

A statistical analysis, using the ANOVA and Turkey–Kramer test methods [18,24], was performed and confirmed that the observed differences in the levels of the PM, CO, UHC and NOx, emissions between the catalyst treated fuels and the reference diesel were considered statistically significant, confirming that the catalyst significantly reduced the exhaust emissions.

4. Conclusions

The exhaust emission characteristics as well as fuel consumptions were investigated using a direct injection diesel engine fuelled with diesel with and without a ferrous picrate-based homogeneous combustion catalyst under various engine operating conditions. Based on the results, the following conclusions can be deduced.

(1) The fuel properties were not altered by the addition of the FTC and FPC catalysts due to their extremely low dosage ratios. Therefore, the catalysts can be used without any engine modifications.
(2) Compared with the reference diesel, the homogeneous combustion catalysts led to decreases in brake specific fuel consumptions under all test conditions. A maximum of 3.7% fuel saving was observed at 3200 rpm engine speed and 0.14 MPa BMEP.
(3) The homogeneous combustion catalysts treated fuels generated remarkably less PM, CO and UHC emissions in the engine exhausts compared with those from the reference diesel fuel. The maximum reduction ratios for PM, CO and UHC emissions were 39.5%, 21.1% and 13.1%, respectively. Slightly elevated NOx emissions were also observed with the application of the ferrous picrate-based catalyst, which was consistent with the improved fuel combustion efficiency and reduced PM, CO and UHC emissions from the test engine.

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