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THE EFFECT OF A HOMOGENEOUS COMBUSTION CATALYST ON THE EMISSION CHARACTERISTICS FROM A COMPRESSION IGNITION ENGINE FUELLED WITH BIODIESEL

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ABSTRACT

This paper reports an investigation into the effect of an iron-based homogeneous combustion catalyst on the emission characteristics from a compression ignition engine fuelled with 100% biodiesel. Prior to the engine tests, the chemical compositions and physical properties of the biodiesel were analysed by using a gas chromatography (GC) and fuel property testers. In the experimentation, the engine was operated under various load conditions at a steady speed of 3200rpm and fuelled with a commercial petroleum diesel (as a reference), biodiesel and catalyst treated biodiesel (1:10000, by volume), respectively. Gaseous emissions (CO, UHC, NOx and CO_2) were measured via an AVL gas analyser and the particulate emissions were evaluated in terms of smoke opacity using a Bosch smoke meter. The results showed lowered smoke, unburned hydrocarbons (UHC) and NOx emissions along with a marginal increase in CO_2 emission for biodiesel, while the CO emission decreased under low loads and increased again under high loads, compared to those for the reference diesel. When operating with catalyst treated biodiesel, it was observed that the smoke, CO and UHC emissions were significantly reduced with slightly increased NOx and CO_2 emissions, which was due primarily to the enhanced fuel combustion by the catalyst. This suggests that the iron-based catalyst is effective in further improving the biodiesel combustion in CI engines.

Keywords: biodiesel, carbon monoxide, compression ignition engine, homogeneous combustion catalyst, smoke emission

1. INTRODUCTION

Compression ignition (CI) engines, also known as diesel engines, are widely used in transportation, heavy machinery and decentralized power generation due to their higher efficiency and durability compared with spark ignition (petrol) engines. However, the increasing demand for diesel engine applications has also led to the increased pollutant emissions, such as unburned hydrocarbons (UHC), carbon monoxide (CO), nitrogen oxides (NOx) and particulate matters (PM), which adversely affect the environment and human health [1, 2]. Recently, under the enforcement of more stringent emission regulations as well as the fast depletion of conventional fossil fuel reserves, the research on alternative fuels with clean combustion in diesel engines has been strongly motivated [3-7].

As a biodegradable, oxygenated and sulphur-free alternative fuel, biodiesel has been extensively studied for its direct application in diesel engines [3, 6, 8, 9]. Biodiesel is a mixture of mono-alkyl esters of fatty acids derived from a wide range of vegetable oils and animal fats and can be mixed with conventional petroleum diesel in any ratio. From the published literature [3, 6, 9], biodiesel generally possesses a higher viscosity, density, flash point, Cetane number and a lower calorific value than conventional petroleum diesel. These differences in the fuel chemical and physical properties between biodiesel and conventional diesel can ultimately affect the fuel combustion efficiency, engine performances and emission characteristics. There is a wide consensus in comprehensive reviews [10-12] that biodiesel combustion tends to reduce CO, UHC and PM emissions at the penalties of slightly elevated NOx and the specific fuel consumption. However, various contradictory findings such as increased CO, UHC or decreased NOx have also been summarised in the referenced review papers which are partly believed to be a consequence of the disparity in fuel chemical structures and the variability in experimental engine conditions [11, 12].

Further improvements in exhaust emissions and fuel efficiencies of biodiesel combustion can be achieved by the application of metal-based catalysts [13-16]. These catalysts consist of a metallic part as the active ingredient that promotes fuel combustion and an organic part to make the catalysts oil soluble, thus the term homogeneous combustion catalysts. There are a number of metals that have been shown to be effective in improving fuel efficiency and reducing exhaust emissions, including platinum (Pt), cerium (Ce), manganese (Mn), magnesium (Mg), molybdenum (Mo), iron (Fe), nickel (Ni), calcium (Ca) and copper (Cu) [13-18]. Gürü et al [14] investigated the influence of Mg and Mo based catalysts on tall oil biodiesel combustion in a CI engine, the test results showed significantly decreased CO and smoke emissions. Kannan et al [15] studied the use of ferric chloride ($FeCl_3$) in waste cooking palm oil biodiesel at the dosage ratio of 20μmol/L. It was observed that, compared to untreated biodiesel fuel, the specific fuel consumption was decreased by 8.6% and the CO, UHC, smoke emissions were reduced by 52.6%, 26.6% and 6.9%, respectively, although with slightly increased NOx emission due to the catalytic effect on biodiesel combustion process.

The authors' previous work [19-22] involved a novel iron-based homogeneous combustion catalyst with ferrous picrate as the active ingredient. Based on our laboratory engine tests [20, 21], the catalyst was observed to save up to 4.2% specific fuel consumption and reduce 33.4% of smoke, 20.5% of CO, 16.2% of UHC emissions when added in a commercial diesel fuel. It was also captured with a higher peak cylinder pressure, heat release rate and a shorter ignition delay induced by the catalyst. However, the application of the aforementioned catalyst in biodiesel combustion in CI engines remains unknown.

The present contribution reports an experimental study of the effect of the ferrous picrate catalyst (FPC) on the emission characteristics of a direct injection diesel engine fuelled with a biodiesel, as a part of our continuing efforts. The exhaust emissions, including smoke, CO, UHC, CO_2 and NOx emissions from baseline reference diesel, biodiesel and FPC treated biodiesel were compared and evaluated under various engine operating conditions.

2. EXPERIMENTAL

2.1 Test engine

The experiments were conducted on a 211cm^3, naturally-aspirated, four-stroke and direct-injection single cylinder diesel engine (YANMAR L48AE-D, AET Ltd.). The main engine specifications are: bore 70mm, stroke 55mm, compression ratio 19.9:1, maximum output 3.5kW at the rated speed of 3600rpm. A water-cooled, 20HP, Zöllner TypeA-100 electric dynamometer was directly coupled to the engine output shaft for providing various load conditions. An automatic controlling system was used to alter the engine speed and output torque. Calibrated sensors/probes were installed to monitor the ambient and intake air temperature, oil temperature, dynamometer coolant temperature and the exhaust temperature. A schematic layout of the diesel engine test system is shown in Figure 1.

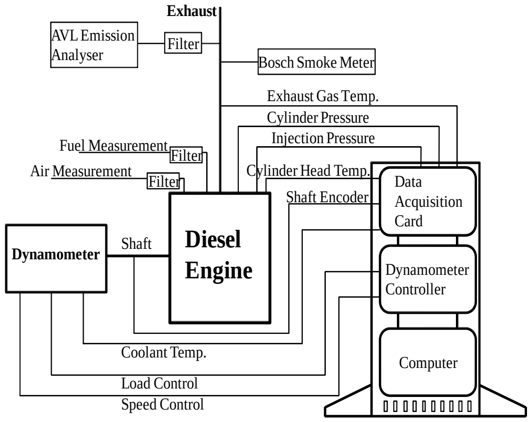

Figure 1 Schematic diagram of the engine test system

For exhaust measurements, an AVL Digas 4000 emission analyser was used to analyse the concentrations of CO, UHC, CO_2 by an infrared method and NOx and O_2 by an electrochemical method. Smoke opacity was measured using a Bosch RTM 430 infrared opacimeter to indicate the overall particulate emissions. To ensure high accuracy of each measurement, the instruments were carefully calibrated at the beginning of the tests. Table 1 shows the accuracies and ranges of the employed instruments for the emission measurements.

Table 1 Specifications of the instruments used for the emission tests

	Measure-ment	Measuring range	Accuracy
AVL Digas 4000	CO	0-10% by vol.	0.01%
	CO_2	0-20% by vol.	0.1%
	UHC	0-20000 ppm vol.	1 ppm
	NOx	0-4000 ppm vol.	1 ppm
	O_2	0-4% by vol.	0.01%
		4-22% by vol.	0.1%
Bosch RTM 430	Opacity	0-100%	0.1%

2.2 Fuels

A commercial diesel (Caltex Australia Ltd.) was used as the reference fuel in this study. The biodiesel was obtained from BioWorks Australia Pty. Ltd. which was derived from the transesterification of soybean oil, rapeseed oil and animal tallow with methanol. The chemical composition of the employed biodiesel was analysed using an Agilent 7890A gas chromatograph (GC) and listed in Table 2. The ferrous picrate homogeneous combustion catalyst (FPC) was provided by Fuel Technology Pty. Ltd. and added into the biodiesel at the ratio of 1:10000 (vol/vol). For convenience, the reference diesel fuel was denoted as "RD", while the 100% biodiesel and FPC treated biodiesel (1:10000) were abbreviated as "B100" and "B100+FPC (1:10000)". The key physical properties of the three fuels were determined and shown in Table 3.

2.3 Test procedure and data collection

The engine was operated under a constant speed of 3200rpm and four load levels, corresponding to the brake mean effective pressure (BMEP) of 0.13MPa, 0.21MPa, 0.33MPa and 0.42MPa, respectively. Under each of the test conditions, the engine was fuelled with "RD", "B100" and "B100+FPC (1:10000)", respectively, and allowed to operate at the specified speed and load for more than 30min until the characteristic temperatures (exhaust, lube oil and coolant) of the engine stabilized. When switching to a different fuel, the engine was purged with the new fuel for at least 30min to eliminate the effect of the fuel in the previous test.

Exhaust emission measurements were taken directly from the exhaust pipe and the data were recorded at 5s intervals for 3min continuous measurements by using a PC with the AVL DiConnect program installed. Each test was repeated three times and the results presented in this paper are the averaged values with the error bars showing the standard deviations. The emission results were normalised on a brake-specific basis (g/kWh) following the procedures recommended by the EPA regulations [23], except for the smoke opacity. Brake specific emission is the mass flow rate of the pollutant divided by the engine power output, which is more useful in reflecting the fuel combustion characteristics [24].

3. RESULTS AND DISCUSSION

3.1 Fuel chemical and physical properties

As shown in Table 2, the chemical composition of the biodiesel employed mainly consists of the methyl esters of palmitic acid (C16:0), oleic acid (C18:1) and linoleic acid (C18:2), at the 18.95%, 43.98% and 23.90% by mass, respectively. The corresponding total saturated and unsaturated fatty acids are 26.53% and 71.76% of the total weight, respectively. Compared to the diesel fuel, the biodiesel has extra oxygen content in its molecules and the large quantity of unsaturated carbon bonds implies high reactivity for combustion.

The main physical properties of the reference diesel, biodiesel with and without FPC catalyst were tested and tabulated in Table 3. Compared with the reference diesel, the biodiesel had a higher viscosity and density, which may affect the fuel injection and atomisation when burning in the engine [11]. A higher flash point was observed with the biodiesel which may indicate a lower volatility. The distillation range of the biodiesel was much narrower than that of diesel because of the initial boiling point (IBP) of biodiesel being higher than that of diesel, in this case, 280°C vs. 177°C. Cetane number of a fuel measures the quality of ignition delay, where higher Cetane fuels have shorter ignition delay period than lower Cetane fuels [11, 12]. The Cetane index of the biodiesel was higher than that of diesel, with 57.1 compared to 50.1, which indicates a shortened ignition delay should be observed when burning the biodiesel in a CI engine.

Table 2 Corresponding fatty acids of the biodiesel esters

Fatty acid	Carbon chain	Formula	Wt%
Myristic	C14:0	$C_{14}H_{28}O_2$	0.95
Palmitic	C16:0	$C_{16}H_{32}O_2$	18.95
Palmitoleic	C16:1	$C_{16}H_{30}O_2$	0.69
Stearic	C18:0	$C_{18}H_{36}O_2$	5.98
Oleic	C18:1	$C_{18}H_{34}O_2$	43.98
Linoleic	C18:2	$C_{18}H_{32}O_2$	23.90
Linolenic	C18:3	$C_{18}H_{30}O_2$	2.78
Arachidic	C20:0	$C_{20}H_{40}O_2$	0.37
Eicosenoic	C20:1	$C_{20}H_{38}O_2$	0.41
Behenic	C22:0	$C_{22}H_{44}O_2$	0.28
Total saturated			26.53
Total unsaturated			71.76
Unknown components			1.71

Note: Cxx:x = No. of carbon atoms: No. of double bonds.

It can also be noted from Table 3 that the addition of the FPC catalyst had no significant effect on the biodiesel fuel properties, as there was no detectable difference in the main physical properties between the pure biodiesel and FPC treated biodiesel. This could be easily understood because of the extremely low dosage ratio (1:10000) of the catalyst used.

3.2 Exhaust emissions

The emission results of the three fuels are presented in Figures 2 to 6, and discussed in the order of smoke, CO, unburned hydrocarbon (UHC), CO_2 and NOx emissions below.

3.2.1 Smoke emissions

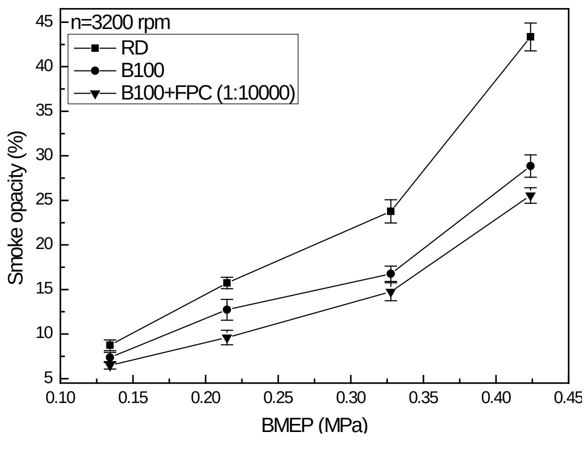

Figure 2 Variation of smoke emissions as a function of engine load for the three fuels at the speed of 3200rpm

Table 3 Specifications of the fuels used in the tests

Parameters	RD	B100	B100+FPC (1:10000)	Analytical Method
Viscosity (cSt at 40 °C)	2.09	3.40	3.40	ASTM D445
Density (kg/m^3 at 15°C)	838.5	883.5	883.2	ASTM D1298
Flash Point (°C)	74	101	100	ASTM D93
Pour Point (°C)	-15	-1	-1	ASTM D97
Sulphur Content (ppm)	<15			ASTM D1266
Distillation Range (°C)	177~360	280~366	279~365	ASTM D86
Calculated Cetane Index (CCI)	50.1	57.1	57.2	ASTM D4737

Smoke emission is a most visually noticeable pollutant from diesel engines which provides an indication of the particulate matters (PM) levels [24]. 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 under oxygen deficiency, low local flame temperature or poor fuel air mixing [24]. It was observed that the smoke opacity from the three fuels increased with increasing in engine load, as shown in Figure 2. This is because at high engine loads, more fuel is supplied to the engine cylinder of a fixed volume (displacement) at each cycle to meet the high power requirement, which in turn affects the mixing of the fuel and combustion air, and therefore, an increase in smoke emission is expected at increased engine loads.

When biodiesel was applied, regardless if the FPC catalyst was added in the fuel or not, the smoke emission was significantly lower under all test points due primarily to the oxygen content in the biodiesel molecule. It was also found a larger decrease in smoke emission for biodiesel under high load conditions, which was consistent with other reports in the literature [8, 25, 26]. The smoke reduction from biodiesel combustion ranged from 11.5~33.4% compared to that of diesel, with the highest ratio achieved at the highest engine load of 0.42MPa BMEP under tested conditions. The observed lower smoke at higher loads from biodiesel was explained in some reports [11, 25] that at high loads, the oxygen content of biodiesel may become more effective in reducing PM especially with high combustion temperature presented in the engine. Another reason could be the absence of aromatics compounds in biodiesel fuels, those being considered as soot precursors, that lead to a dramatic smoke reduction [12, 27].

It was also observed that the smoke emission was further suppressed by the addition of the FPC catalyst to the biodiesel under all tested conditions. Compared to the untreated biodiesel, the smoke reduction ratio for the FPC treated biodiesel was from 11.5~24.4%, while it was from 22.1~41.1% when compared to that of the reference diesel. Further to our previous findings of the effectiveness of the FPC catalyst in enhancing combustion process and reducing smoke emission from diesel fuel [20, 22], this suggests a possible extended application of the FPC catalyst for biodiesel application in diesel engines.

3.2.2 CO emissions

CO emission is considered as a consequence of incomplete combustion of the fuel in diesel engines, especially when oxygen in local combustion area is insufficient, short residence time and/or the combustion temperature is low [24]. Figure 3 plots the variation of the brake specific CO emissions from the three fuels versus engine load. All the three CO curves show a similar overall trend, which is: from low to medium load conditions (0.13~0.33MPa BMEP), the CO emission tended to decrease with increasing the engine load due to improved fuel efficiency and increased combustion temperature; from medium to high loads (0.33~0.42MPa BMEP), CO emission increased again because in this load range, in order to meet the high power output, an increased amount of fuel was injected to the engine at each cycle which might result in poor fuel-air mixing, shortened residence time and therefore an increase in the CO emission.

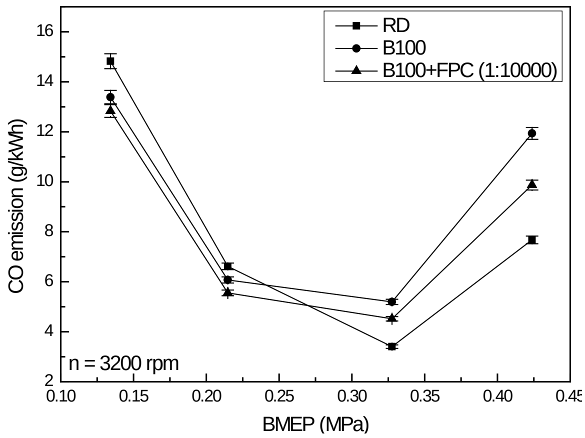

Figure 3 Variation of CO emissions as a function of engine load for the three fuels at the speed of 3200rpm

The CO emission level from the biodiesel was observed to be lower than that of the reference diesel under the low and medium load conditions, which is easily explained by the extra oxygen content present in the biodiesel and thus more complete combustion than the petroleum diesel [11]. However, the CO emission was deteriorated from the load of 0.33MPa BMEP upwards for the biodiesel fuel. This can be explained as the air-fuel mixing process was affected by the difficulty in biodiesel atmomisation because of its high viscosity and density, together with the relatively less air available when more fuel is injected to the engine cylinder at the high loads, thus the resulting locally fuel-rich mixtures of the biodiesel may have led to more CO formation during combustion [12, 27].

The addition of the FPC in biodiesel was effective in reducing CO emissions, as lowered CO was observed from Figure 3 in the whole test load range, compared to that of the biodiesel. Meanwhile, the trend of decreasing CO from the FPC treated biodiesel was more obvious with increasing engine load, with the reduction ratio ranging from 4.2~17.3%, compared with the untreated biodiesel. Similar to the mechanism proposed in our previous work on improved diesel combustion [20-22], the FPC acts as a combustion catalyst to significantly improve the efficiency of biodiesel combustion in the CI engines and is again proved to be capable of further reducing the incomplete combustion products from the combustion of biodiesel fuels.

3.2.3 UHC emissions

Unburned hydrocarbon (UHC) emission from diesel engines is another parameter for determining the fuel combustion inefficiency [24]. The general trend of the UHC emissions from the three fuels was to decrease with increasing engine load, as depicted in Figure 4. This is because of the improved fuel combustion efficiency, high fuel thermal efficiency and high flame temperature achieved under high loads [21, 22].

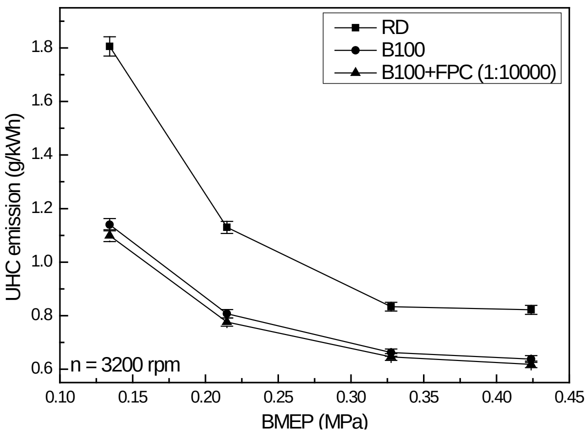

Figure 4 Variation of UHC emissions as a function of engine load for the three fuels at the speed of 3200rpm

The biodiesel (B100) exhibited drastically lowered UHC emissions, with the reduction ratio ranging from 20.6~36.9% under the engine conditions tested. This behavior was observed to be more noticeable at lower engine loads. Reasons for lowered UHC emission from biodiesel, as reviewed by Lapuerta et al [11], could be: (1) the higher oxygen content in biodiesel molecules which leads to more complete combustion; (2) the higher Cetane number which reduces the ignition delay and relates to the UHC decrease; and (3) the advanced ignition timing which associates with a lower UHC formation. It was also noticed that lowered UHC emissions of biodiesel were obtained under all loads, unlike the CO emissions, which were increased again from the medium to high loads. This was explained by some authors [11, 12, 28] that during biodiesel combustion, UHC was first converted to CO and then to CO_2 provided sufficient oxygen was available in the local combustion zone. Further to the discussion in section 3.2.2, the results of UHC emissions may add another explanation for the deteriorated CO emissions under high load conditions, that the oxidation of UHC to CO consumed most of the oxygen, leaving less oxygen available for the reaction of CO to CO_2, and all the reasons added together led to the higher CO emissions (at higher loads) observed in the present study.

A further decrease in UHC emission was found from the FPC treated biodiesel, although it was quite slight, with the reduction ratio ranging from 2.5~3.8% compared to that of the untreated biodiesel and from 22.5~39.2% compared to that of the reference diesel. This is consistent with the observations of reduced smoke and CO emissions by the use of the FPC catalyst in biodiesel, which again implies the improved efficiency of biodiesel combustion in the CI engines.

3.2.4 CO_2 emissions

CO_2 is a normal product of combustion of hydrocarbon fuels and the high CO_2 concentration in exhaust emissions is an indication of more complete combustion of the fuel [12]. From Figure 5, it was found that the CO_2 emissions from all the three fuels decreased with increasing engine load, which was due to the improved brake thermal efficiency and the reduced brake specific fuel consumption under high loads [21]. Compared to the reference diesel, a slightly higher CO_2 emission was observed from the biodiesel combustion under all test conditions. The increased CO_2 is considered to be due to the more complete combustion and the increased fuel consumption rate because of the lower heating value of biodiesel fuel [11, 12].

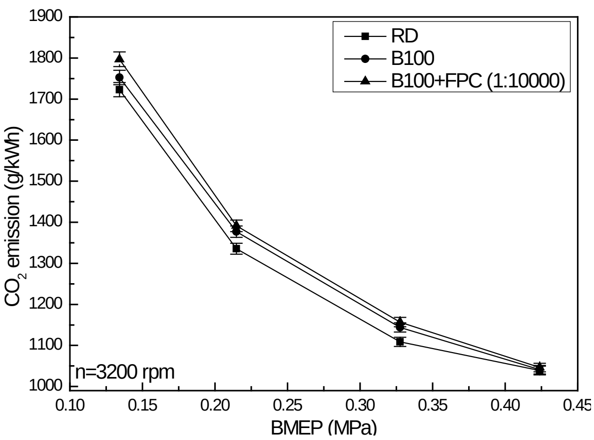

Figure 5 Variation of CO_2 emissions as a function of engine load for the three fuels at the speed of 3200rpm

More CO_2 emission was observed when adding the FPC catalyst in the biodiesel, with the ratio increasing from 0.5~2.5% compared to that of the untreated biodiesel and 0.7~4.3% compared to that of the reference diesel, under the engine conditions tested. Correlating to the acquired results of the FPC catalyst on reducing smoke, CO, UHC emissions from biodiesel combustion, the slight increase in CO_2 emission is reasonable and acts as an evidence to reveal the improved efficiency of biodiesel combustion in the presence of the FPC catalyst.

3.2.5 NOx emissions

The formation of nitrogen oxides (NOx) is mainly governed by the peak flame temperature, the local oxygen concentration and the combustion duration [24]. Figure 6 shows that, regardless of the operating modes, specific NOx emissions from the biodiesel were slightly less than that of the reference diesel, ranging from 4.5~8.6%. The decrease in NOx emissions with biodiesel has also been reported by many others [6, 15, 25, 28-30]. The reasons, as reviewed by Lapuerta et al [11] and Xue et al [12], were believed to be: the fuel physical properties of biodiesel (higher density and viscosity, lower volatility etc.) may have influenced fuel injection and evaporation, and consequently, the NOx formation; the higher Cetane number of the biodiesel would result in a shortened ignition delay, allowing less time for the fuel-air mixing before the premixed burning phase, therefore, leading to a lower thermal NOx formation. As discussed above, the formation of NOx from biodiesel combustion in CI engines is very complex, as this is not quantitatively determined by a single change in the fuel properties but rather a result of a number of interacting mechanisms, whose effects may tend to reinforce or weaken under different engine conditions and specific fuel characteristics [11, 12].

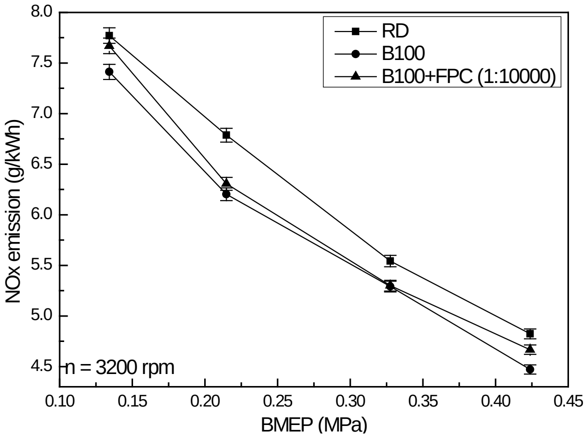

Figure 6 Variation of NOx emissions as a function of engine load for the three fuels at the speed of 3200rpm

When the FPC catalyst was applied to the biodiesel, the NOx emissions were slightly elevated, as observed from Figure 6, being 0.2~4.5% higher than that of the biodiesel. This is consistent with other reports [13-15] when applying metal-based catalysts in biodiesel fuels. The increased NOx emissions is considered to be a result of the improved combustion by the addition of the FPC catalyst, which is reasonable by considering the aforementioned reductions in incomplete combustion products, i.e. smoke, CO and UHC emissions.

4. CONCLUSIONS

A systematic study has been carried out to investigate the effectiveness of the ferrous picrate homogeneous combustion catalyst on the emission characteristics of a compression ignition engine burning a biodiesel under various engine operating conditions. From the experimental results, the following conclusions can be deduced:

Compared to the reference diesel, the biodiesel possessed a higher viscosity, density, flash point, pour point, Cetane index and a narrower distillation range with higher initial boiling point (IBP). The addition of the FPC catalyst had no noticeable effect on the physical properties of the biodiesel due to the extremely low dosage ratio (1:10000, vol/vol).

Compared to the reference diesel, the biodiesel combustion emitted lower smoke, unburned hydrocarbon (UHC) and NOx because of its extra oxygen content and high Cetane number. CO emission was decreased in the low load range and then deteriorated in the high load range due primarily to the poor fuel-air mixing and locally fuel-rich mixtures of biodiesel. Higher CO_2 emission was also observed as a consequence of the more complete combustion and the increased fuel consumption rate of the biodiesel burning in CI engines.

The smoke, CO and UHC emissions from the biodiesel combustion were further decreased by the addition of the FPC catalyst, with the highest reduction ratios up to 24.4%, 17.3% and 3.8%, respectively, compared to those of the untreated biodiesel. The slight increases in NOx and CO_2 emissions were also observed from the FPC treated biodiesel which may result from the improved efficiency of the biodiesel combustion in the presence of the FPC catalyst.

The addition of the FPC catalyst is effective in improving biodiesel fuel combustion in the CI engines and capable of significantly suppressing the incomplete combustion products from burning biodiesel fuel.

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