

Experimental Studies On Reduction of Nox Emission of Biodiesel Fuelled Engine Using A Novel Natural Additive

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Abstract

The world's energy demand increases because of increase in human population. The conventional fossil energy resources will be depleted and burning of these conventional fuels increases greenhouse gases and causes global warming. These problems can be overcome using renewable alternative energy resources such as biofuels. In recent years biodiesel is considered as a renewable alternative to the fossil diesel. The utilisation of biodiesel as a fuel in compression ignition engine results in lower CO, HC and Smoke emissions as the biodiesel is an oxygenated fuel. The major problem with the biodiesel is its higher NO_x emission and different techniques were used to minimise NO_x emission. The use of synthetic and nano-metal additives to the biodiesel may affect the environment and hence it is necessary to identify non-toxic, low cost, biodegradable, and sustainable additives to reduce the NO_x emission. Hence an attempt was made to use clove oil as a natural additive for the honge oil biodiesel as it has better antioxidant property. The engine tests were conducted with various dosages of clove oil such as 1000 and 2000 ppm and the engine load were increased with an increment of 25% up to full load. The addition of clove oil to the honge oil biodiesel significantly affects the engine NO_x and clove oil can be substituted as an additive to reduce the NO_x emission of the biodiesel fuelled CI engine and without altering the engine hardware. The clove oil enhances the oxidation stability of the honge oil biodiesel.

Introduction

The fossil fuels consumption is rapidly increasing due to industrialisation and increase in energy demand of the human population. The use of fossil fuels for energy generation increases greenhouse gas emissions. Hence many researchers carrying out research to find renewable fuels which are CO₂ neutral. The energy is prime mover which drives the social and economic developments of any country. The depletion of fossil fuels has focussed research on development of sustainable alternative renewable energy sources. The biomass-based energy sources are considered as alternative to the petroleum fuels [1]. Hence importance is given to research work focused on bio-fuel development and adoption in recent years. The biodiesel can be used as partial substitute for the diesel [2]. The exhaust gases of the diesel engine cause significant air pollution which directly and indirectly affect the environment. The use of biodiesel as partial substitute significantly reduces the engine emissions [3]. The biodiesel can be used as a partial replacement for the diesel to avoid food versus fuel related issues [4]. The future technological development should concentrate on utilisation of non-edible and low-cost feed stocks for biodiesel production and newer biodiesel production technologies to reduce the production cost [5]. Transesterification method is used to produce biodiesel from fats and vegetable oils. The biodiesel produced by alternative energy stimulant methods satisfies the biodiesel standards and minimise the biodiesel production time and reduces the production cost [6].

India's is one of the developing countries and its surface transport dependent on fossil oil import. Hence India's bio-fuel policy recommends usage of biofuel as partial substitute for the fossil diesel and petrol. In India, non-edible oils are chosen as feedstock to produce biodiesel and hence the non-edible oil seed

plants are grown in waste and dry lands [7]. In the transportation sector, India's fuel consumption may double by 2030 [8]. The honge biodiesel is considered as replacement to diesel and biodiesel can be obtained with honge with a yield of 99% at the optimised reaction condition. The B20 honge biodiesel diesel blend gives engine performance comparable to diesel [9]. Also the honge biodiesel blend minimise the engine emissions such as CO and PM, except NOx [10]. The composition of the biodiesel affects the fuel oxidation during combustion [11]. The biodiesel is an oxygenated fuel contains reactive radicals. These radicals enhance the diffusion combustion, increase the combustion speed and minimise the combustion duration. Hence the combustion temperature of the biodiesel is higher and causes higher NOx level as compared to the fossil diesel [12]. The impact of the oxygenated fuels on each polycyclic aromatic hydrocarbon derivative may be different from each other [13].

The biodiesel properties are similar to diesel; however, the biodiesel has lower volatility and lower oxidation stability. Hence antioxidant additives, oxygenated additives, metallic and non-metallic based additives are added to the biodiesel to enhances its fuel properties and to get better engine performance and lower emissions. The synthetic antioxidants, propyl gallate (PG), butylated hydroxyanisole (BHA), butylated hydroxytoluene (BHT), etc are added to the biodiesel to enhance its oxidation stability. The addition of 15% of diethyl ether (DEE) as an oxygenated fuel additive to the biodiesel blend increases the brake thermal efficiency (BTE) and reduces the brake specific fuel consumption (BSFC). Also, it increases the net heat release rate compared to the diesel [14]. The use of oxygenated additive, DEE, reduces the NO and CO emissions and slightly increases the HC emission [15]. Hence DEE can be blended with fossil diesel and biodiesel to minimise the engine emissions.

The biodiesel fuelled compression ignition engines emits higher NOx emission. Hence it is necessary to reduce the NOx emission of the CI engine using suitable methods or using additives. Presently few methods such as adopting emulsion of water and diesel as fuel, direct water injection into the combustion chamber and adding water into the inlet manifold as these methods reduce combustion chamber temperature [16, 17] are used to reduce the NOx emission. Also the engine NOx emission can be controlled by retarding injection timing, lower injection pressure and using exhaust gas recirculation (EGR) method [18]. The addition of nanoparticle such as TiO₂ reduces engine emissions such as CO and HC. It is also reported that the addition of TiO₂ with EGR results in lower emission and better performance of the engine [19]. The synthetic antioxidant PG enhances the oxidation stability of the honge oil biodiesel. Also, it significantly reduces the NOx emissions of the CI engine with BTE like the diesel [20]. The synthetic antioxidants possess greater risks of side effects [21]. The synthetic antioxidant additives such as BHA and BHT may be toxic to the biological systems. Few natural additives (example, basil oil) have better antimicrobial and better antioxidant [22]. The natural additives are nontoxic, biodegradable, and eco-friendly and hence these additives are better replacement to commercially available synthetic additives [23]. The oxidation stability of biodiesel can be enhanced by adding small dosage of bio-oil [24]. The ginger extract can be used as an antioxidant for the pongamia biodiesel and the addition of 250 ppm to the biodiesel increases the oxidation stability and the biodiesel meets both American and European biodiesel

oxidation standard specifications [25]. The addition of nano-additives influences the emissions and performance of biodiesel fuelled CI engine. The magnetic nanoparticles added to the rice bran biodiesel minimise the emission levels of CO, HC and NO_x [26]. The SFC, CO and UHC of the CI engine can be minimised by adding graphene oxide nano-particles to the *Ailanthus altissima* biodiesel blends. However, the NO_x emission increases [27]. The urea-SCR system reduces the NO_x emission of B20 blend of waste cooking oil biodiesel and diesel. The addition of cobalt oxide and manganese oxide nano-particles to the waste frying oil biodiesel blend (B20) enhances the bsfc and also minimises both NO_x and CO emissions [28]. The use of Al₂O₃ nanoparticle to the biodiesel blend reduces the HC emission by 62% and CO emission by 12% at 50% engine load condition increases the NO_x emission by 10% [29]. The use of L-ascorbic acid in *Borassus flabellifer* oil reduces NO_x, smoke, HC and CO [30]. The use of the hydrazides as additive do not affect the engine's NO_x emissions [31]. The EGR method minimise the NO_x level, however this method increases the HC, CO and smoke levels. The EGR with the use of TiO₂ nanoparticle minimises the BSFC, HC and CO levels. Hence the combination of EGR and nanoparticle is one of the effective methods in improving engine performance and minimizing the engine emissions [32].

The assessment studies of the toxicological effects of nanoparticles on environment and human health are minimum. Hence the research work must be carried out to study the mechanisms of toxicity of nanoparticles [33]. The large-scale use of nanoparticle may result in major health concern [34]. Hence it is necessary to identify non-toxic, biodegradable, and low-cost bio additives to enhance the fuel properties of the biodiesel and also to minimise the engine emissions. It is reported that the bio-based additives adversely affect few fuel properties of biodiesels, however the variation are small and in acceptable range. The *Pongamia pinnata* leaves extract increase the BTE and minimises the CO, HC and NO_x [35]. The experimental work shows that the natural additives such as L-Ascorbic Acid 6-palmitate, caffeic acid and tannic acid improve the viscosity and oxidative stability [36]. The addition of bio-oil additive reduces NO_x level and smoke opacity of the biodiesel. However, CO and HC values increases and these emissions levels lower than the diesel [37]. The *T. Cordifolia* stem extract contains higher amount of total phenolic content which has good free radical scavenging activity. It enhances the oxidation stability of the biodiesel and it is reported that this natural extract can be used as low cost, non-toxic, eco-friendly natural antioxidants [38]. A study was carried out to study the antioxidant properties of natural extracts derived from hibiscus flowers, senna leaves and blackberry fruits shows that the senna leaves has better properties [39]. The *Melia azedarach*, *Psidium guajava* and *Albizia lebbek* (AL) are used as natural leaf extract additive for the B20 jamun biodiesel blend. The effectiveness of antioxidants properties at 1000 ppm dosage with the biodiesel blend is *AlbiziaLebbek* > *Melia Azedarach* > *Psidium Guajava*. The engine study results shows that the addition of 1000 ppm of AL to the biodiesel blend minimises the NO_x as compared to fossil diesel [40].

The honge is a fast-growing tree that grows in the semiarid regions India. The honge trees are commercially cultivated and grow in different types of soils. The colour of the honge oil is yellow or reddish-brown and its heating value is approximately 41 MJ/kg. The oil is nonedible and acrimonious in

taste^[41]. The potential for honge oil to produce biodiesel is high in India, however it has lower oxidation stability. Hence antioxidants are added with the honge oil biodiesel. Hence in this work, we have tried clove oil as a natural antioxidant for the honge oil biodiesel and studied the influence of clove oil on emissions and performance of the engine. Figures 1 and 2 shows the honge tree and honge seeds.

The clove is the median size tree, and it is one of the important spices. It is used for preservation foods and used in medicinal applications. The flower buds of clove are pale in color finally turn into bright-red clove buds. The clove oil contains large amount of phenolic compounds such as eugenol, gallic acid and eugenol acetate. It has better antioxidant and antimicrobial activity. The clove oil has excellent free radical scavenging activity and inhibit hydroxyl radicals^[43]. Figure 3 shows the clove seed.

Materials And Methods

The acid value of non-edible honge oil was high and hence a two-step transesterification was used to produce biodiesel. In the first step, the acid value of the honge oil was reduced acid catalyst esterification reaction. In the second step, biodiesel was produced using base catalyst transesterification. The products of the transesterification are biodiesel and glycerine, and these products are allowed to settle for three hours and biodiesel was separated from the glycerine. The raw biodiesel was subjected to water wash to remove the impurities. After water wash, the moisture present in the biodiesel was removed by heating it above 100° C. The properties of the honge oil biodiesel were determined as per ASTM procedures.

The honge oil biodiesel has lower oxidation stability and hence we used clove oil as natural antioxidant. Also, the effect of honge oil biodiesel on engine fuelled with honge oil biodiesel was studied. The clove oil was added with the concentration of 1000 and 2000 ppm to the honge oil biodiesel. The clove oil completely dissolves in the honge oil biodiesel. As per the ASTM procedure, properties of the biodiesel added with clove oil and biodiesel were measured.

A single cylinder compression ignition engine was retrofitted to modify it as experimental setup to carryout engine tests. A dynamometer of eddy current type was used to change load of the engine. The suitable instrumentations were used to record important observations. The details of the experimental setup of the engine are shown in the Table 1. Table 2 shows the details of the engine exhaust gas analyser and error analysis is shown in the Table 3. The AVL make emission analyser was used to measure the engine's emission level. A thermocouple was used to measure the temperature of the exhaust gases. The engine tests were performed at steady state condition and the engine load was increased from 0 to 100% with an increment of 25%. Initially engine tests were performed with fossil diesel to get baseline engine test data. Further engine tests were performed with biodiesel and biodiesel added with clove oil. Figure 4 (a) depicts the schematic of engine test setup and 4(b) shows photo of the engine test setup.

Table 1
Details of the engine experimental setup

Engine	Compression ignition engine
Number of cylinder	One
Engine Coolant	Water
Make	Kirloskar
Cylinder Displacement	661 CC
Engine Capacity	5.2 kW
Rated Speed	1500 rpm
Compression Ratio of Engine	17.5 : 1
Stroke and Bore of Engine	110 mm and 87.5 mm
Fuel Injection Type	Direct injection
Loading Device	Eddy Current Dynamometer
Type of Thermocouple Used	K – Type, Range : 0 to 1200 Degree C

Table 2
Exhaust gas analyser principle and measuring range

Gas Analyser		
Type of Emission	Working Principle	Measuring Range of Gas Analyser
CO.	NDIR	0 to 2.0 % vol
HC.	NDIR	0 to 10000 ppm
NOx.	Electrochemical	0 to 5000 ppm
Smoke meter Make :AVL and Model : 437C		
Smoke Opacity of smoke meter		0.00 to 100 %

Table 3
Instruments accuracy and uncertainty

Instrument Name	Instrument Accuracy	% Uncertainty of instrument
Fuel measuring unit (cc)	± 0.10	± 0.200
stopwatch (sec)	± 0.60	±0.030
Speed measuring unit (rpm)	± 10.00	± 0.100
Thermocouple (°C)	± 1.00	± 0.110
Load cell (kg)	± 0.10	± 0.100

Results And Discussion

Table 3 compares the properties of the fuels used in this work. BC1000 and BC2000 indicate the clove concentration of 1000 and 2000 ppm respectively. The biodiesel has higher flash and fire point than the diesel. Hence it is easy to store, transport and handle the biodiesel. However, BC 1000 has lower flash point as compared to the biodiesel. The addition of clove oil slightly reduces the flash and fire points. The density of the biodiesel increases with clove oil concentration. The oxidation stability of the honge biodiesel significantly increases with increase in clove oil concentrations. This is because the clove oil has good free radical scavenging activity. The pour point of the biodiesel added with clove oil is better than the biodiesel. However, the clove oil does not affect the viscosity and calorific values significantly.

Table 3
Comparison of Important Fuel Properties of the fuels

Property	Diesel	Biodiesel	BC1000	BC2000
Fuel Flash Point (°C)	65	161	152	146
Fuel Fire Point (°C)	71	130	148	141
Fuel Density at 15°C (kg/m ³)	862	894	884	880
Fuel Oxidation Stability (h)	-	3.5	10.8	11.5
Fuel Pour Point (°C)	-13	3	2	1
Calorific Value of Fuel(MJ/kg)	42.71	39.41	39.29	39.05
Fuel Viscosity at 40°C (mm ² /s)	2.64	3.94	3.91	3.84

Table 4
Comparison of important fuel properties of the fuels

Property	Diesel	Biodiesel	BC1000	BC2000
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The engine performance tests were carried out on the engine experimental setup with different types of fuels. The impact of clove oil on performance and emissions of the CI engine are discussed below. The term brake thermal efficiency (BTE) indicates the performance of the engine and it shows the effectiveness of energy conversion from the fuel into engine shaft power. The impact of the clove oil on BTE is shown in the Fig. 5. The BTE gradually increases with increase in load. A slight difference in BTE was observed with different fuels at low at part loads. The BTE of diesel is higher than the other fuels at all loads. The BTE of the biodiesel is lower than diesel due to higher flash point and poor volatility of the biodiesel. The addition of clove oil to the biodiesel slightly increases the BTE at higher load. This is due to better volatility and flash point of the clove oil which enhances the spray formation and better premixed combustion. The clove oil of 2000 ppm causes lower BTE as compared to the biodiesel at 100% load.

The quality of the combustion is indicated by the engine combustion temperature and the engine exhaust gas temperature (EGT). The impact of fuels on EGT at different loads is depicted in the Fig. 6. The EGT value increases with increase in engine loads as the fuel consumption increases gradually with increase in load. The higher fuel consumption increases the combustion temperature and EGT. The variation in EGT at low load is small for BC1000 and BC2000; however, the variation is significant for diesel and biodiesel. The biodiesel results in higher EGT than the diesel due to higher viscosity and lower volatility of the biodiesel which results in slower combustion. The slower burning of biodiesel in the diffusion combustion phase increases EGT. The clove oil reduces the EGT of the biodiesel due to free radical scavenging activity of the clove oil. This process lowers combustion temperature and lower EGT. The EGT of BC1000 is higher than the BC2000.

The incomplete combustion of the fuel inside the combustion chamber of the engine results in CO emission. The CO emission is high at full load due to insufficient air supply, low flame temperature and fuel rich zones ^[42]. The impact of fuels on engine CO at various load is shown in the Fig. 7. The honge oil biodiesel is an oxygenated fuel and contains reactive radicals. These radicals improve the diffusion combustion, increase the combustion speed and reduce the combustion duration. Hence the CO emission

of honge oil biodiesel is lower than diesel. The CO emission of the engine increases with the addition of the clove oil to the biodiesel. The hydroxyl (OH) radicals promote the oxidation of CO into CO₂. The clove oil contains phenolic compounds which has better antioxidant activity^[43] and results in excellent free radical scavenging activity and inhibits the formation of hydroxyl radicals^[44]. The reduction in concentration of hydroxyl radicals reduces the oxidation of CO^[45]. The CO emission of the BC1000 is higher than the honge oil biodiesel and lower than the diesel. The CO emission of the BC2000 is higher than BC1000 as the BC2000 significantly reduces the availability of hydroxyl radicals required for oxidation of CO.

The influence of different fuels on engine's un-burnt hydrocarbon (HC) emission at various load is illustrated in the Fig. 8. The HC level of the engine is affected by the fuel properties, fuel atomization, spray formation and combustion efficiency. Since the honge oil biodiesel is an oxygenated fuel that enhances the flame oxidation and results in higher flame speed, particularly in the fuel-rich regions. This causes enhancement of the oxidation of unburned HC which reduces the HC emission^[46]. The BC1000 and BC2000 results in higher HC emission as compared to the honge oil biodiesel. This is due to the free radical scavenging activities of the clove oil. However, BC 1000 gives lower HC emission as compared to BC2000 and diesel. The BC2000 has good radical scavenging activity and reduces the OH radical formation which inhibits the HC oxidation. Hence BC2000 results in higher HC emission. Similar type of behaviour of the antioxidant is reported in the literature^[47, 48].

The impact of various fuels on oxides of nitrogen (NO_x) emission of the engine at various engine loads is depicted in the Fig. 9. The NO_x is one of the major emissions of the CI engine. The reason for NO_x emission of the engine is due to higher combustion temperature, higher oxygen concentration and higher residence time of the fuel inside the combustion chamber. The honge oil biodiesel emits higher NO_x emission due to higher bulk modulus of the honge oil biodiesel which slightly advances the fuel injection timing and increases the residence timing of the fuel inside the combustion chamber. The biodiesel is an oxygenated fuel and hence it also increases the combustion temperature and NO_x emission. The BC1000 results in lower NO_x level as compared to honge oil biodiesel. The clove oil contains eugenol which has excellent radical scavenging activity^[49]. The addition of clove oil reduces the generation of hydroxyl radicals during combustion of the fuel. The reduction in hydroxyl radicals during combustion hinders the combustion and reduces the combustion temperature as well as NO_x emission. This result is similar to the studies reported by other researchers^[50]. Hence, clove oil can be used as additive to reduce the NO_x level.

The variation in smoke opacity of the engine at different engine loads is illustrated in Fig. 10. The smoke is one of major emission emitted by the compression ignition engine and it is visible part of the emission. The smoke emission is formed due to poor mixing of fuel and air, insufficient residence time and insufficient oxygen for the combustion of fuel. The smoke emission of the diesel is higher than all the fuel. The smoke emission of honge oil biodiesel is low as the biodiesel is an oxygenated fuel which enhances the oxidation of the fuel and reduces smoke formation. The smoke emission of BC2000 is

higher than the smoke emission of BC1000 and biodiesel. The addition of clove oil increases the reductive reactions and inhibits the oxidative reactions and quenches the free radicals during the combustion, which could increase the emission of smoke emission^[50]. The clove oil contains phenolic compounds which can scavenge the alkylperoxyl radical. Hence the smoke emission of the BC1000 and BC2000 are more than the honge oil biodiesel. The smoke emission of the biodiesel BC1000 and BC2000 are lower than the diesel.

Conclusion

The presence of unsaturated components reduces the oxidation stability of honge oil biodiesel. The clove oil addition to the honge oil biodiesel improves its oxidation stability. The addition of clove oil to the honge oil biodiesel impacts the engine's NOx emission significantly. However, there is a slight increase in CO, HC and smoke emissions. This is because of free radical scavenging activities of the clove oil which minimises the oxidation and inhibits the combustion of the fuel. However, the increase in CO, HC and smoke levels are lower than the diesel. The addition of clove oil to the honge oil biodiesel does not have much influence on the thermal efficiency of the engine. Hence the clove oil can be used as an antioxidant to the honge oil biodiesel with reduction in NOx emission significantly.

Declarations

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Availability of data and materials

The data related to this work is available within manuscript.

Authors 'contributions

N.Kapilan : Conceptualisation, metholdogy, experimentation, funding acquisition, R.P.Reddy : Drafting, analysis, investigation, Abu Saleh Ahmed: visualization, review, editing

Ethics approval and consent to participate.

Consent for publication

All authors have given consent for publication.

Competing interests

We declare that we do not have known competing financial interests that may appear to influence the work reported.

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Figures



Figure 1

Honge Tree



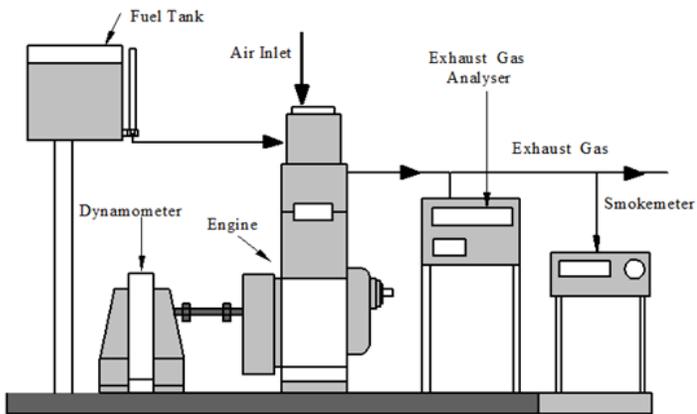
Figure 2

Seeds of Honge



Figure 3

Clove Seed



A

B

Figure 4

(a) Schematic of the experimental setup (b) Engine Experimental setup

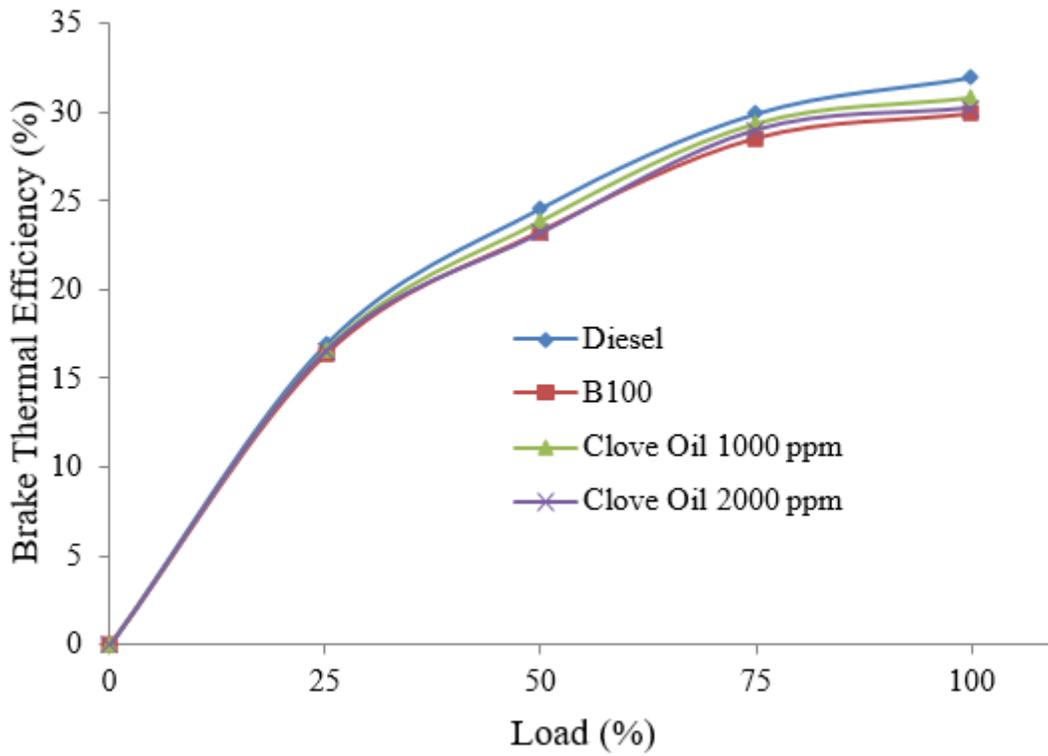


Figure 5

Brake Thermal Efficiency at various Engine loads

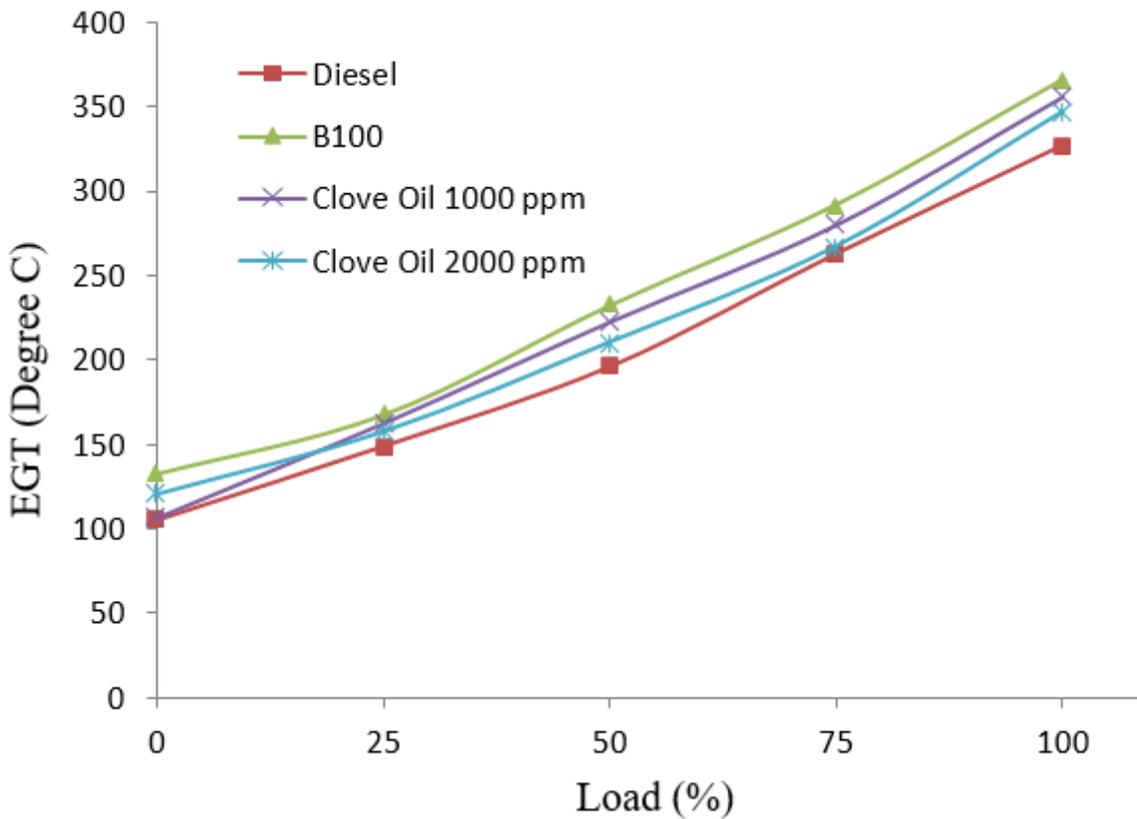


Figure 6

EGT at various Engine loads

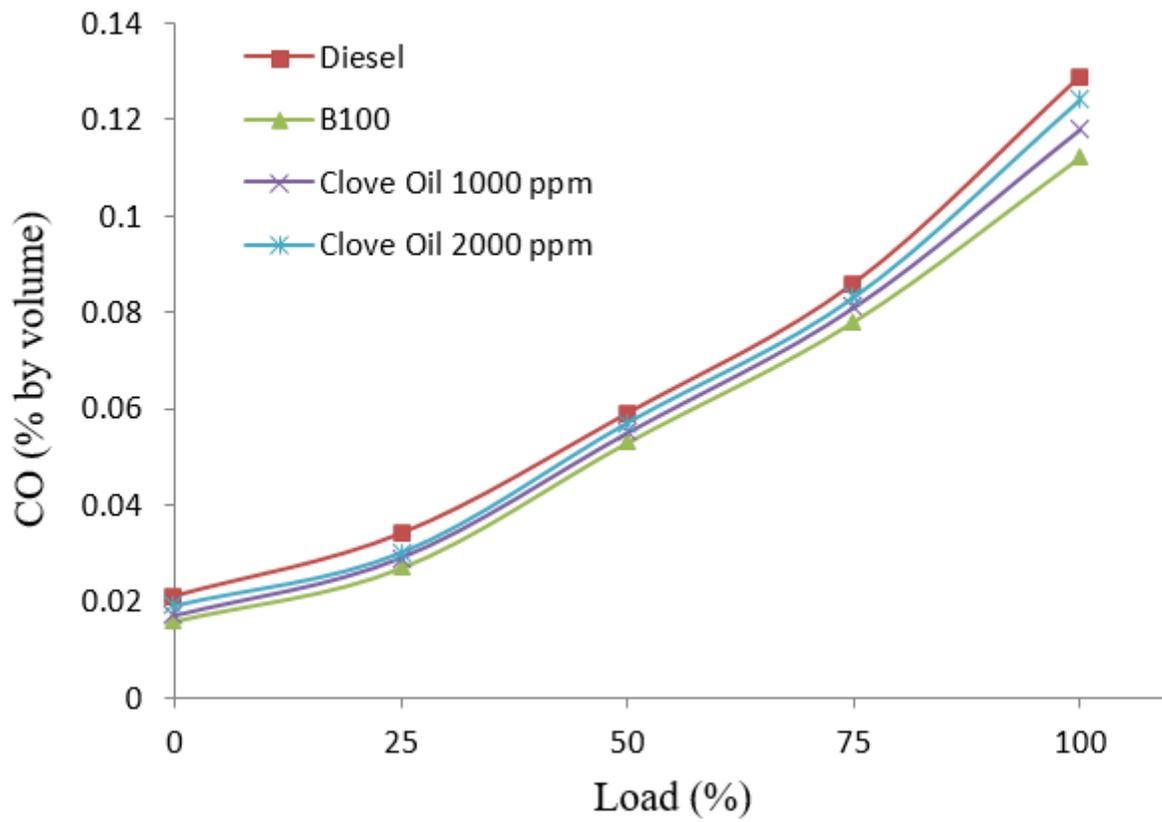


Figure 7

CO at various Engine Loads

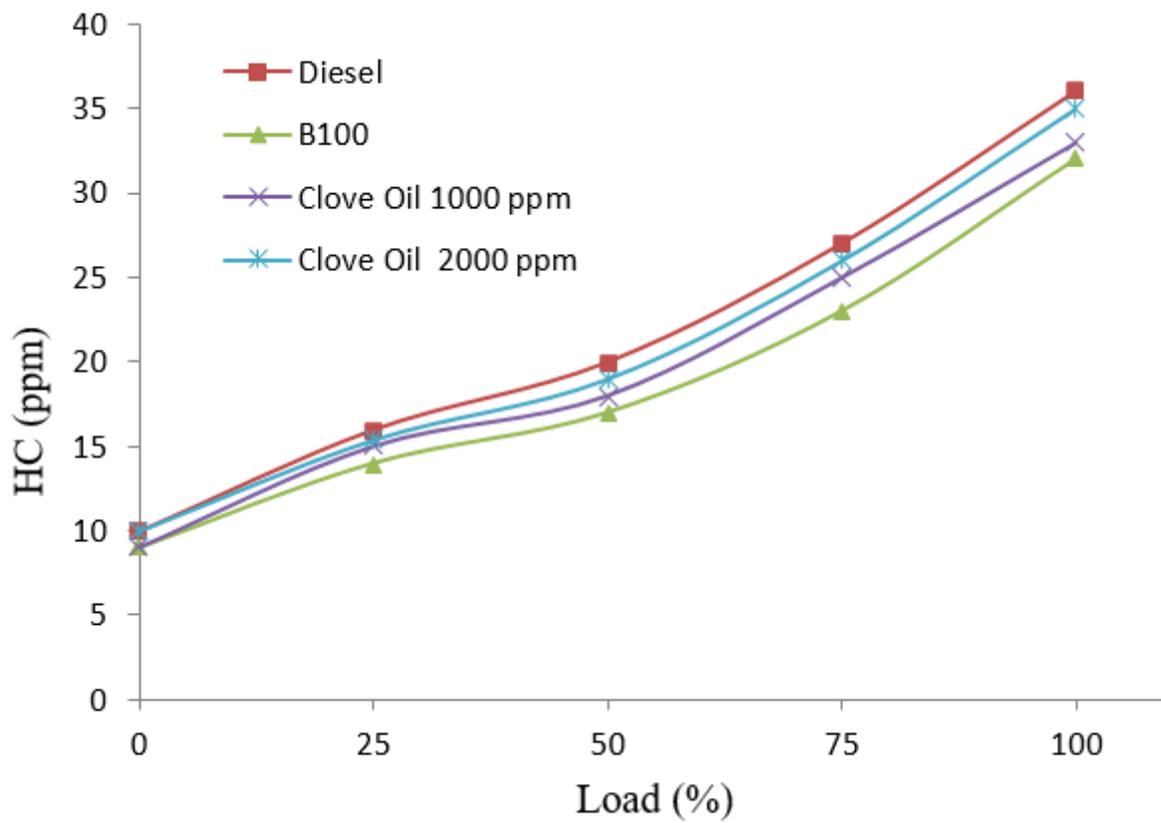


Figure 8

HC at various Engine Loads

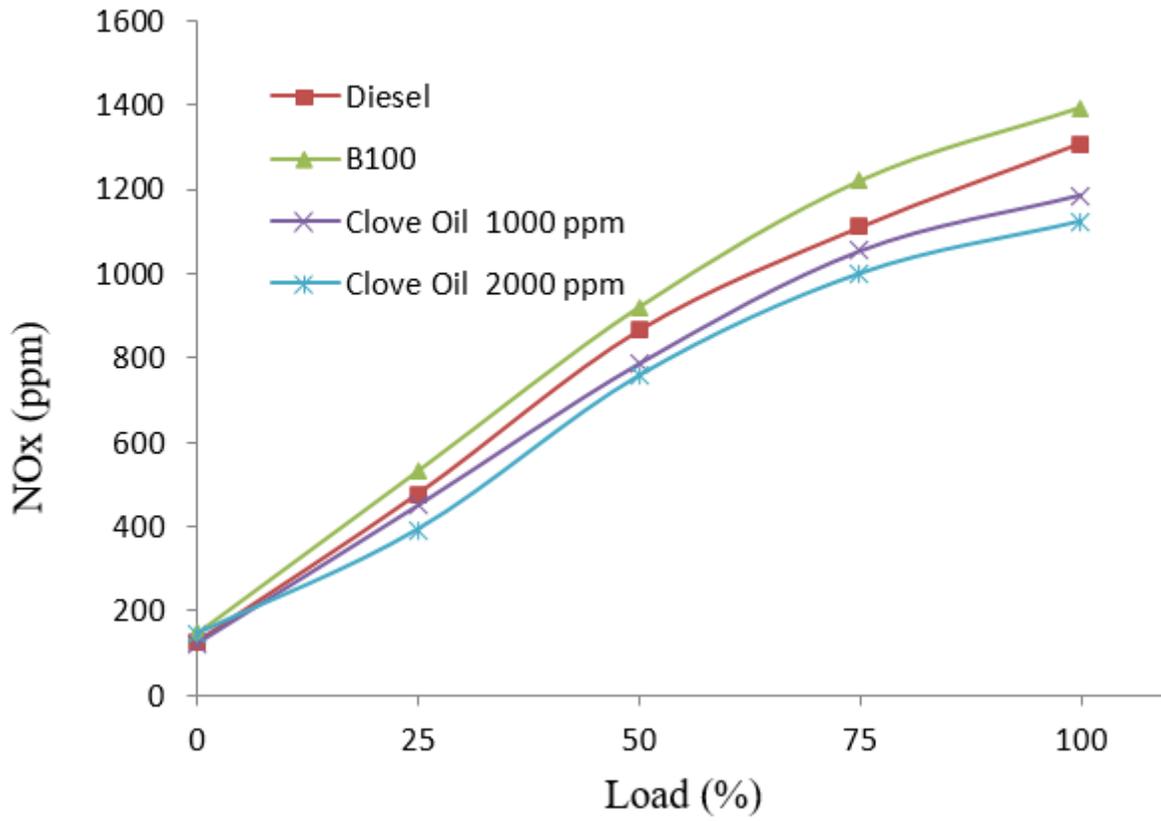


Figure 9

NOx at various Engine Loads

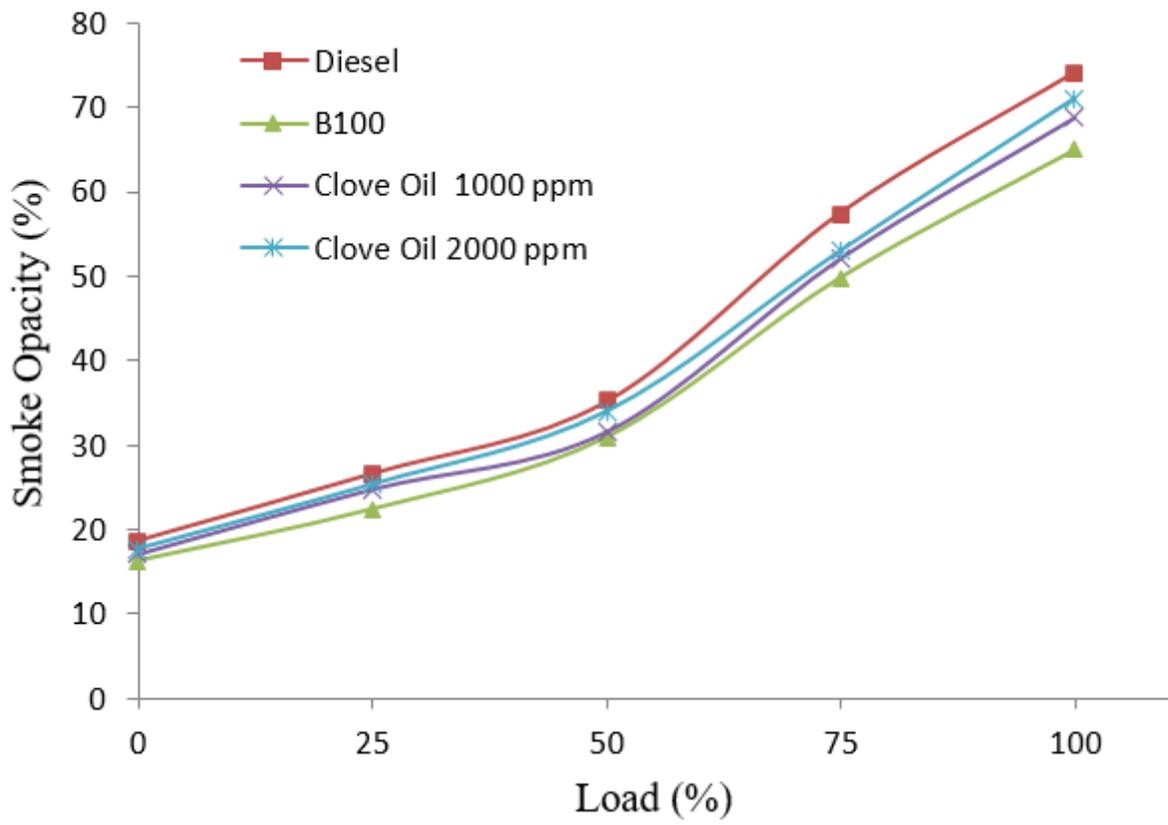


Figure 10

Variation in smoke emission at various Engine Loads