

EMISSION REDUCTION OF METHANOL GASOLINE BLENDED FUELS IN LEAN BURN SI ENGINE USING ZEOLITE-BASED CATALYTIC CONVERTOR

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ABSTRACT

Because of the high thermal efficiency and low fuel consumption, the Gasoline Direct Injection (GDI) engines are preferred by the Automobile manufacturers. Researchers worldwide are focussing to reduce our reliance upon fossil fuel based products and for a cleaner energy environment through the alternative source of fuel. As it is not possible to replace the fossil fuel, we can reduce the usage of biofuels by blending of the gasoline with methanol. Methanol addition in the GDI engine will increase the overall oxygen availability inside the combustion chamber and thus improving the combustion process. As the temperature is increased the formation of the NO_x emission will also increase from the engine. In order to reduce the excess NO_x emissions, a specially fabricated metal doped zeolite based catalytic convertor has been used. The GDI engine was tested for different blends of methanol-gasoline namely M0 (methanol 0% and gasoline 100%), M5 (methanol 5% and gasoline 95%) and M10 (methanol 10% and gasoline 90%) fitted with commercial catalytic convertor and the zeolite based catalytic convertor for the various speeds of the engine rpm (2000, 2200, 2400, 2600 and 2800). The performance characteristics of different methanol-gasoline blends were analysed and the emission reducing capacity of commercial as well as Cu-ZSM5 coated catalytic convertor were analysed for all the blends. From the results it can be concluded that the M10 blend is having better brake thermal efficiency (BTE) and lesser unburned HC and CO emissions.

KEY WORDS : Cu-ZSM5 Zeolite, GDI engine, ZSM5, SI engine, NO_x reduction, Methanol-gasoline blend.

INTRODUCTION

The crude oil demand is keep on increasing especially in the countries like china and India which are the major consumers of crude oil to meet its energy requirements (Sadala *et al.*, 2019). Most of the crude oil energy in the world were spend for the powering the transport and Industrial sectors. This crude oil based Internal Combustion (IC) engines produces various harmful emissions in the environment (Elavarasan, 2019). The major emissions from the IC engines are Hydrocarbons (HC), Carbon Monoxide (CO) and Nitrogen Oxides (NO_x) (Rajasekar *et al.*, 2010). To overcome these oil depletion, some countries shifting towards

alternative source of energy. Methanol is also a prominent source of the alternative fuel which can be blended with the petrol and can be used in Spark Ignition (SI) Engine (Kumar and Tomar, 2019). This blended methanol fuel requires no modification for lower volumetric composition (Mourad and Mahmoud, 2019). The presence of methanol in the gasoline will increase the anti-knock properties and the brake thermal efficiency of the fuel (Wang *et al.*, 2019). Also the presence of the methanol will increase the oxygen content of the fuel and thus reducing the HC and CO emissions (Yang *et al.*, 2019b). Zhang *et al.* (2019) have observed that, if the alcohol blend was increased, then the compression ratio of the engine also should be increased in

proportion to the blend in gasoline (Zhang and Sarathy, 2016). Zaharin *et al.* (2018), have also confirmed that the testing of the iso-butanol additive in the alcohol blend will not require any modification of the SI engine (Zaharin *et al.*, 2018). Thakur *et al.* (2018) have recorded that the maximum brake thermal efficiency were recorded when the 20% of alcohol-gasoline blend was used for testing (Thakur *et al.*, 2017). Iodice *et al.*, (2018) have found that ethanol-gasoline blends of 20% can overcome the cold starting problem of an SI engine (Iodice, Langella and Amoresano, 2018). Even for 20% of the ethanol-gasoline blends, there is no modification required in the engine (Fan *et al.*, 2019). But the increase in the concentration of methanol in gasoline will increase the emissions and also the results in the uneven combustion inside the engine (Biswal *et al.*, 2019). In this research we have used various methanol-gasoline blends in a lean burn SI engine and the emission results were tested in commercially available catalytic convertor and a metal doped zeolite based catalytic convertor. The methanol concentration in the gasoline is having good characteristics of converting the unburned HC to H₂O and CO to CO₂, but it will increase the formation of the NO_x. The NO_x formation in the combustion chamber is mainly due to the reaction of the oxygen and the nitrogen at higher temperature. The availability of the oxygen in a lean burn GDI engine is usually more, the addition of methanol content in the mixture will also increase the oxygen availability, this is the reason for the improved combustion process in a methanol blended gasoline fuel (Chang and Park, 2019). Bacariza *et al.* (2019), have already proved that the ZSM5 coated monolith in the exhaust stream have better tendency to reduce NO_x emission (Bacariza *et al.*, 2019). The NO_x reduction capacity of the ZSM5 zeolite can be further increased by an added active catalyst metal in it (Balzarotti *et al.*, 2019). Copper, Iron and Cerium were some of the low cost metals that can be used in the ZSM5 zeolite as an active catalyst metal for the reduction of emissions in the exhaust (Ko *et al.*, 2019). The results of the NO_x emission reduction can be even increased by adding one more metal in the ZSM5 zeolite coated monolith (Azarikhah *et al.*, 2019). As the ZSM5 zeolite can be obtained from the waste fly ash material from the thermal power station and the catalytic materials were non-precious metal, the cost of the metal doped zeolite catalytic convertor is very much lesser than of the commercial catalytic convertor (Lin *et al.*, 2019). In this study, we

have used Cu-ZSM5 type coated monolith for the reduction of the NO_x emissions and the results were compared with the commercial catalytic convertor.

Fuel Preparation

Three types of fuels were considered for testing in the lean burn SI engines. The methanol and gasoline were mixed at three different volume rates were prepared for testing and it was shown in the Table 1 (Godwin *et al.*, 2019).

Table 1. Composition of methanol and gasoline used for testing.

| S. No. | Sample Name | Volume of Methanol (Vol %) | Volume of gasoline (Vol %) |
|--------|-------------|----------------------------|----------------------------|
| 1 | M0 | 00 | 100 |
| 2 | M5 | 05 | 95 |
| 3 | M10 | 10 | 90 |

A. Measurement of Physiochemical Properties

The properties for each fuel blend and the gasoline were presented in the Table 2 (Schifter *et al.*, 2011).

Catalyst Preparation

ZSM5 zeolites were bought from and zeolite manufacturing company named zeolyst International from USA and Cupric Chloride was bought from Fisher scientific from India. The ZSM5 zeolite was taken as the base material the CuCl₂ was taken as the transition material preparation. The Cu-ZSM5 zeolite was prepared by Na⁺ ion exchange method for the coating in the monolith for the NO_x emission reduction.

B. Na⁺ ION Exchange Method

Because of its simplicity the Na⁺ ion exchange method was considered for the preparation of the Cu-ZSM5 zeolite. The purchased 100 g ZSM5 was taken and added with the 100 mL of 0.5M CuCl₂ and the whole solution was poured into a beaker containing 1000 mL distilled water. The solution is allowed to stirred using a magnetic stirrer for about 16 hours in normal room temperature for the ion exchange reaction to takes place between Na and Cu (Yuan *et al.*, 2018). Then this mixture is filtered and washed with the distilled water to remove the free ions and it was placed in a furnace at 500 °C for 6 hours to get the final product Cu-ZSM5 (Yoldi *et al.*, 2019).

Table 2. Some Properties of gasoline and methanol-gasoline blends

| Properties | M0 | M5 | M10 | Methanol |
|---|--------|--------|--------|----------|
| Net Heating Value (MJ/Kg) | 42.600 | 41.462 | 40.268 | 19.850 |
| Density (kg/m ³ at 15 °C) | 750.8 | 751.9 | 754.1 | 796 |
| Kinematic Viscosity (mm ² /s at 40 °C) | 0.494 | 0.529 | 0.548 | 0.596 |
| Research Octane Number | 95 | - | - | 108.7 |
| Motor Octane Number | 85 | - | - | 88.6 |
| Latent Heat of Vaporization (kJ/kg) | -380 | - | - | 1185 |
| Auto Ignition Temperature (°C) | -370 | - | - | 423 |
| Distillation (%) | | | | |
| Initial Boiling point | 45 | 64 | 42 | 43 |
| 30 | 71 | 64 | 58 | 55 |
| 60 | 113 | 64 | 110 | 108 |
| 90 | 168 | 65 | 163 | 165 |
| End Boiling Point | 207 | 66 | 207 | 206 |
| Recovery (%) | 96 | 99.2 | 96.4 | 96.4 |

C. X-ray Fluorescents (XRF)

The chemical components present in the ZSM5 zeolite and the prepared Cu-ZSM5 zeolite was identified by XRF method using Philips Spectrometer PW1401. The results obtained for both the commercial ZSM5 zeolite and the prepared Cu-ZSM5 zeolite were shown in the Table 3. It can be observed from the table that the percentage of sodium oxide was decreased and the percentage of Copper was increased in the Cu-ZSM5 type zeolite. This is because of the ion exchange happened between Cu and Na during the Na⁺ ion exchange method.

Table 3. % Mass of Zsm5, Cu-Zsm5

| Composition | ZSM5 | Cu-ZSM5 |
|--------------------------------|--------|---------|
| SiO ₂ | 88.372 | 87.012 |
| Al ₂ O ₃ | 3.256 | 3.091 |
| Fe ₂ O ₃ | 0.114 | 0.110 |
| CaO | 0.014 | 0.014 |
| MgO | 1.150 | 1.050 |
| SO ₃ | 0.134 | 0.126 |
| Na ₂ O | 7.001 | 1.515 |
| K ₂ O | 0.00 | 0.00 |
| CuO | 0.00 | 5.012 |
| P ₂ O ₅ | 0.00 | 0.00 |
| TiO ₂ | 0.025 | 0.22 |
| BaO | 0.00 | 0.00 |
| LOi | 0.00 | 0.00 |

D. Cordierite Monolith

The Oxidation monoliths and some blank monoliths for the coating of Cu-ZSM5 zeolite were purchased from Bo cent Advanced Ceramics Co Ltd, China for the preparation of after treatment device. The

monoliths have 400 cpsi and cell density with 0.17 mm wall thickness and the diameter and the length of the monolith were equal size of 90 mm. The exhaust gas was initially allowed to pass through the oxidation monolith and then to the Cu-ZSM5 zeolite. The oxidation monolith can help to convert CO to CO₂ and HC to H₂O and the Cu-ZSM5 coated monolith can help to reduce the NO_x emission to N₂ and O₂. For the preparation of Cu-ZSM5 zeolite coated monoliths. Figure 1 shows the picture of blank monoliths.

**Fig. 1.** Picture of Blank Monoliths

E. Monolithic Washcoating

Because of the effectiveness and the simplicity of operation, Dip coating method was adapted for the coating of the Cu-ZSM5 zeolite to the monolith. The initial step was the preparation of the slurry having 50% of Cu-ZSM5, 4% of silica gel and remaining 46% of distilled water. The weight of the blank monolith should be identified initially with the help

of a weighing machine. The Blank monoliths were made to immerse the slurry completely for 2 minutes. Then the monoliths were blown with the compressed air from one end to other to remove the excess sticky slurry and it was placed in a closed type muffle furnace for about 2 hours at 130 °C. Again the same process from immersing the monolith in the slurry was repeated till the weight of the deposition reaches 16% weight of the monolith. After this, the monolith is Calcined at 500 °C for about 5 hours (Mehla *et al.*, 2019).

F. Fabrication of Catalytic Converter

The Oxidation monolith and laboratory made Cu-ZSM5 zeolite coated monolith were placed in a steel casing and then fitted with the exhaust stream of the engine. The photographic view of the Cu-ZSM5 coated catalytic converter in the engine was shown in the Figure 2 and the commercial catalytic converter is shown in the Figure 3.

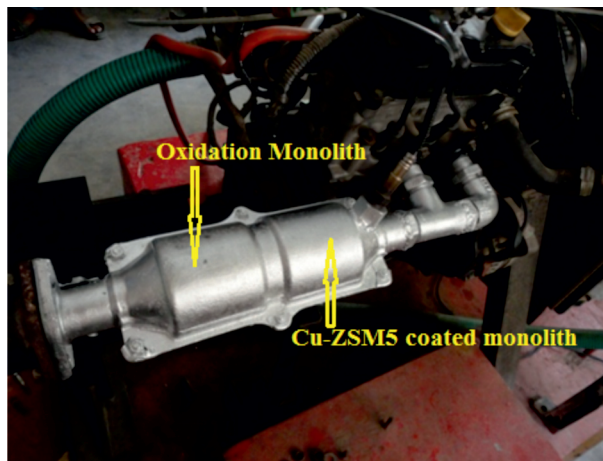


Fig. 2. Picture of Cu-ZSM5 coated catalytic converter

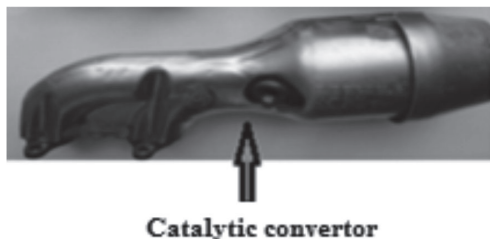


Fig. 3. Picture of Commercial catalytic converter

Experimental Apparatus and Procedure

The lean burn SI engine was connected with an eddy current dynamometer was used for the conduct of the experiment. The schematic view of the experimental setup is shown in Fig. 4. The

experiment procedure consists of testing of the three types of methanol-gasoline (M0, M5, M10) blends in the SI engine at different speed conditions and evaluating the performance and emission characteristics for each blend. The emission values are observed with the help of the AVL 444,a 5 gas analyser. Then the two methanol-gasoline blends (M5, M10) were taken for the testing to evaluate the emission reduction capacity of commercial catalytic converter and with Cu-ZSM5 coated catalytic converter. In all the conditions the engine was allowed to reach the steady state and then the emission values are noted in the gas analyser. The general specifications of the test engine are given in Table 4.

Table 4. Specification of Petrol Engine

| Type | SI Engine |
|---------------------|--------------------------|
| Number of cylinders | 2 |
| Displacement | 624 cc |
| Bore | 73.5 mm |
| Stroke | 73.5 mm |
| Compression Ratio | 9.5:1 |
| Fuel | Petrol |
| Cycle | 4-Stroke |
| Max. Engine output | 25.74 kW @ 5250 rpm |
| Max. Torque | 48 Nm @ 3000 rpm |
| Orifice Diameter | 20 mm |
| Cooling System | Water |
| Loading Device | Eddy current dynamometer |

The experiments are conducted at different engine speeds ranging from 2200 to 3000 rpm by 200rpm increments, and at each of the engine speeds 5 different fuels were tested. The fuel blends were shown in Table 1. The emission generated from these fuels were recorded initially without attaching the catalytic converter, secondly with OEM catalytic converter and finally within house made Cu-ZSM5 catalytic converter. A total of 30 experiments were conducted. Engine brake power exhaust gas temperature, fuel consumption and emissions such as CO, HC, CO₂, O₂ and NO_x were measured during the experiments. For each experiment, the engine was allowed to reach a steady state condition and then the measurements were recorded in a few seconds. No data was taken until brake power and speed were maintained at 1% of the fluctuation.

RESULTS AND DISCUSSION

G. Brake Thermal Efficiency

The variation of the various blends of the methanol-

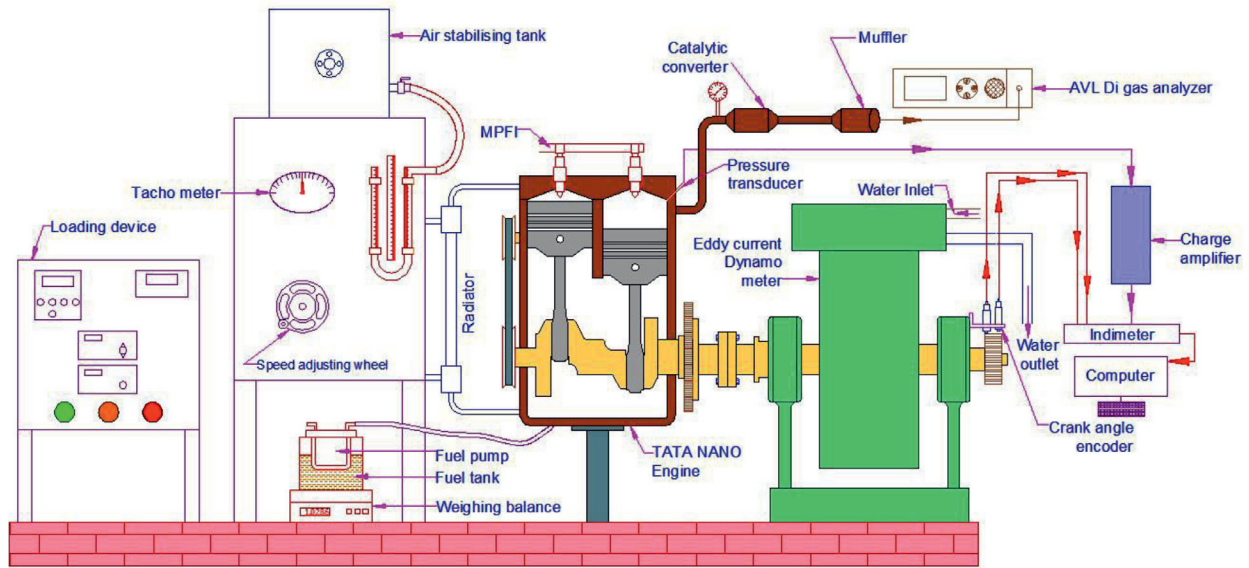


Fig. 4. Schematic Diagram of Experimental setup

gasoline blends without any catalytic converter against the engine speed was shown in the Figure 5. The M10 blend has the maximum BTE compared to the M5 and M0, the reason is that the laminar propagation of the M10 blend is higher than that of the other blends (Yin *et al.* 2016). Thus the combustion process is enhanced with the increase in methanol concentration in the blends. It is observed from the figure that the BTE of the blends M5 and M10 at 2400rpm were 3%, and 6% higher than that of the M0 fuel.

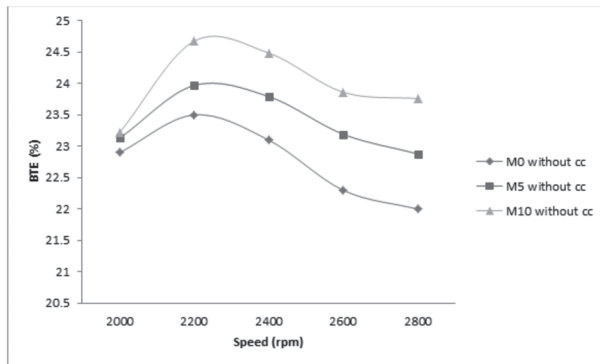


Fig. 5. BTE of various test fuel against engine speed

H. CO Emissions

Figure 6 shows the variation of CO emissions of various methanol-gasoline blends against the engine speed. At 2600rpm, the CO emissions of M5 and M10 were 6% & 10 lesser than that of the M0 fuel. CO emission is formed mainly due to the deficiency of oxygen in the combustion chamber (Yang *et al.*,

2019c). Compare to M0, M5 and M10 have more oxygen in it and results in the oxidation of the CO to CO₂.

The efficiency of the commercial catalytic converter and the Cu-ZSM5 zeolite coated catalytic converter for the oxidation of the CO emissions formed from M5 and M10 blends was performed.

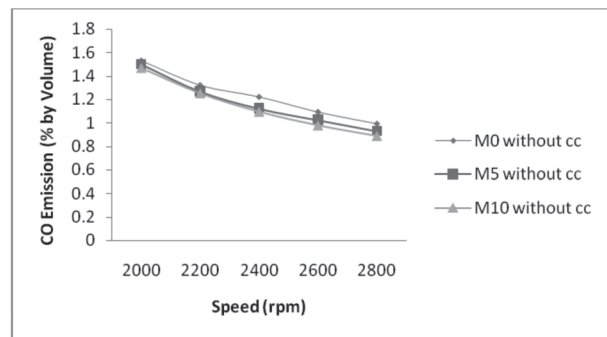


Fig. 6. CO emission of various test fuel against engine speed

The corresponding graph is shown in the Figure 7. It can be observed from the figure that the commercial as well as Cu-ZSM5 catalytic converters reduces the CO emission from the blends, but the zeolite based catalytic converter have the maximum efficiency. At 2400 rpm, the values of CO emissions observed by the gas analyser for various blends with the commercial catalytic converter is 40% lesser than that of the Cu-ZSM5 coated catalytic converter. This is due to the conversion process of CO to CO₂ occurs both in the oxidation monolith as well as Cu-ZSM5 monolith (Liu *et al.*, 2018).

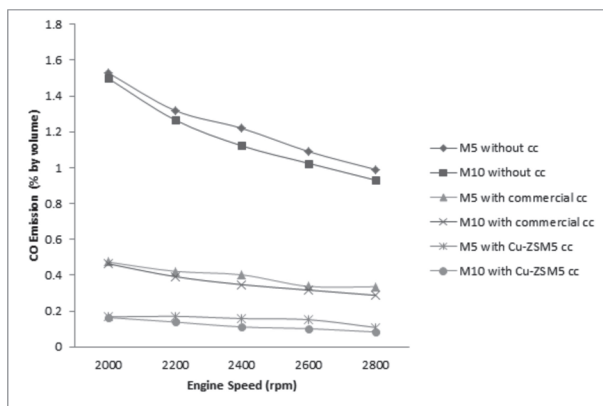


Fig. 7. CO emission of test fuels with catalytic converters against engine speed

I. HC Emissions

The HC emission formation is decreasing with the increase in the speed for all the methanol-gasoline blends. From the Figure 8 it can be observed that there is a decrease of about 10% and 12% of HC emission formation in the blends M5 and M10 at 2400 rpm when compared with M0. The reason for the reduction of the emission with increase in the speed is due to the reduction of the flame quenching in the combustion chamber and also the temperature of the engine increases that will reduce the unburned HC (Yang *et al.*, 2019a).

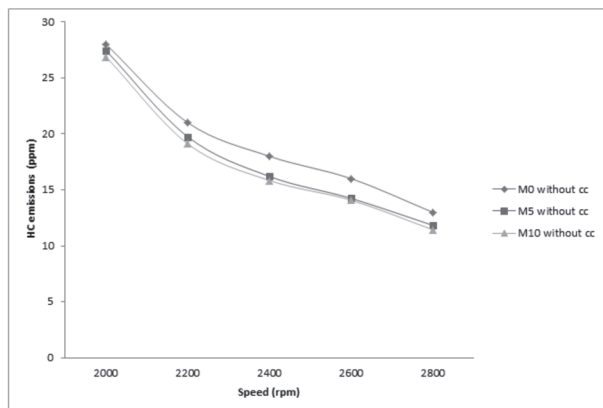


Fig. 8. HC emission of various test fuel against engine speed

The efficiency of the commercial catalytic convertor and the Cu-ZSM5 zeolite coated catalytic convertor for the oxidation of the HC emissions formed from M5 and M10 blends was performed. The corresponding graph is shown in the Figure 9. It is observed from the figure that the efficiency of the oxidation of HC emissions for Cu-ZSM5 coated zeolite catalytic convertor was better than that of the commercial catalytic convertor. At 2400 rpm, HC

oxidation in the Cu-ZSM5 coated catalytic convertor is 39.3% more than that of the commercial catalytic convertor. This is because of the availability of the oxygen in the Cu-ZSM5 zeolite is more due to the dissociation of the NO_x to N_2 and O_2 (Gallastegi-Villa *et al.*, 2016). Thus the oxidation of HC takes place in both the oxidation monolith and the Cu-ZSM5 coated monolith inside the catalytic convertor results in maximum oxidation of HC compared to the commercial catalytic convertor.

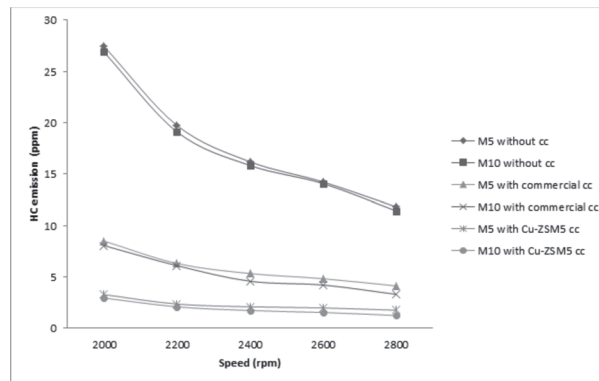


Fig. 9. HC emission of test fuels with catalytic converters against engine speed

J. NO_x Emissions

The NO_x emission formation is increasing with the increase in the speed for all the methanol-gasoline blends. From the Figure. 10 it can be observed that there is an increase of about 7% and 17.3% of NO_x emission formation in the blends M5 and M10 at 2400rpm when compared with M0. The reason for the high emission with increase in the speed is due to the complete combustion and the rise in temperature of the engine increases the formation of the NO_x emissions (Zhang *et al.* 2019).

The efficiency of the commercial catalytic

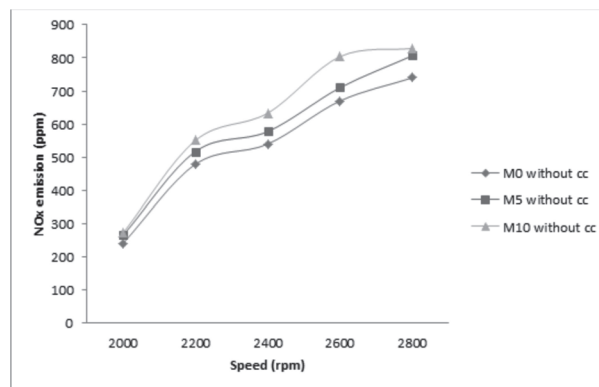


Fig. 10. NO_x emission of various test fuel against engine speed

converter and the Cu-ZSM5 zeolite coated catalytic converter for the reduction of the NO_x emissions formed from M5 and M10 blends was performed. The corresponding graph is shown in the Figure 11. It is observed from the figure that the efficiency of the reduction of NO_x emissions for Cu-ZSM5 coated zeolite catalytic converter was better than that of the commercial catalytic converter. At 2400 rpm, the NO_x reduction in the Cu-ZSM5 coated catalytic converter is 28.16% more than that of the commercial catalytic converter. This is because of the activity of the Cu-ZSM5 zeolite in the monolith, there will be more active sites available for the disassociation of the NO_x (Martinez-Hernandez *et al.*, 2015).

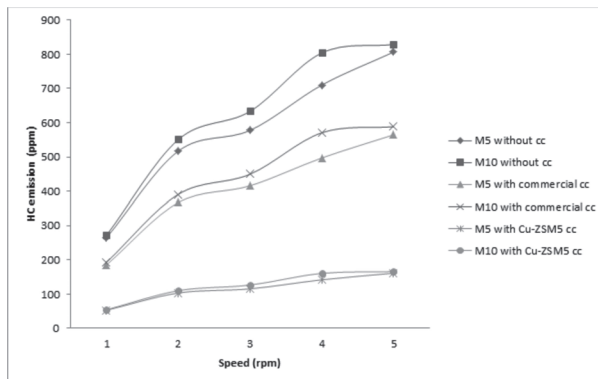


Fig. 11. NO_x emission of test fuels with catalytic converters against engine speed

CONCLUSION

- The Cu-ZSM5 zeolite has been successfully tested for the emission reduction of the M5 and M10 methanol-gasoline blend over a wide range of speed.
- NO_x conversion efficiency achieved by Cu-ZSM5 zeolite coated converter is 28.16% more effective than the commercial catalytic converter for all the speeds.
- The CO conversion efficiency achieved by Cu-ZSM5 coated zeolite catalytic converter 40% more than the commercial catalytic converter.
- The HC conversion efficiency achieved by Cu-ZSM5 coated zeolite catalytic converter is 39.3% more than that of the commercial catalytic converter.
- Back pressure developed is well within the acceptable limits across the catalytic bed.
- No appreciable deactivation of converter was observed, even after 100 hours of experimental examination of zeolite based catalytic converter.

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