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The Perspectives of Diesel Exhaust Emission: Effects and Control of NOx Emission from Diesel Engine

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***Abstract*— Exhaust emissions from Diesel vehicles are now a day's more concern. Diesel vehicles emit more amount of NOx than the petrol driven vehicles. Due to implementation of stringent emission norms BS IV, it is the demand to develop a catalytic converter which shall reduce the emission considerably. This paper summarizes the problem and solutions of NOx emission from diesel vehicles.**

In this paper an experimental investigation of two designs (Cylindrical and Convergent-Divergent shape) of diesel catalytic converter in the Jeep exhaust gas stream is reported. Two transition metal oxide catalysts (5% Ag and 5% Fe) loaded over alumina support were filled in these catalytic converters and heated externally by means of a heating tape upto 2800C.

In 5% Silver loaded catalyst, the maximum NO conversion of 36.36% achieved at temperature of 2800C under full acceleration in the cylindrical catalytic converter and 31.41% conversion with Convergent-divergent catalytic converter. When 5% Iron catalyst was tested in Cylindrical catalytic converter the NO conversion was 27.08% and with Convergent-divergent catalytic converter it was 25.25% at 2800C.

Transition metal oxide based catalytic converter containing 5% Silver and Iron catalyst is an efficient system to reduce the NOx emission from diesel exhaust. Introduction of new future fuels will also be able to reduce NOx emissions in near future.

Keywords-Vehicular exhaust pollution; NOx pollution; Emission norms; Diesel catalytic converter; Transition metal catalysts.

I. INTRODUCTION

In India the transport sector is receiving fast growth due to increase in population and living standard of people. There is a remarkably increase in diesel fuel operated Public utility vehicles (PUV) and Special utility vehicles (SUV). The diesel fuel operated vehicles emit more exhaust pollutants as compared to the petrol operated vehicles.

All these are increasing exhaust pollution like hydrocarbon, carbon monoxide, NOx & SOx in the local environment. NOx emissions are having serious environmental concern because of their role in smog formation. NOx being more hazardous, the limit is set to 350 ppm in many countries, but the emission from diesel engines are generally up to 2500 ppm. For heavy duty diesel engines of BS IV standard (Bharat standard IV) the CPC (Central pollution control board) of India has set the limit of Nitric Oxide emission as 3.5 g/kW-h. As per National ambient air quality standard the NOx concentration upper limit for residential / industrial and village area is 80 micro gram/ m^3 of air. The implementation of Indian emission norms for diesel operated vehicles is given in Table-1.

Table 1: Implementation of Indian emission norms for diesel engines

Year	Emission limit in g/kW-h			
	CO	HC	NOx	PM
1991 norms	14.0	3.5	18.0	-
1996 norms	11.2	2.4	14.4	-
India stage 2000 norms	4.5	1.1	8.0	0.36
2002- Bharat stage II	4.0	1.1	7.0	0.15
2005- Bharat stage III	2.1	1.6	5.0	0.10
2010- Bharat stage IV	1.5	0.96	3.5	0.02

The pollutants emitted by diesel engines are CO, NOx, Hydrocarbons, aldehydes, ketones, soot, smoke & odour. The NOx emission levels of diesel engines greatly vary according to engine power rates. Engines having larger bores, lower speed and higher power rates have higher NOx levels in general. Typical constituents of diesel exhaust gases are as shown in Table-2.

Table 2: Constituents of Diesel engine exhaust gases

Major constituents (more than 1%)	Minor constituents (less than 1%)
Water (H_2O) Carbon dioxide (CO_2) Nitrogen (N_2) Oxygen (O_2)	Oxides of sulphur (SO_2, SO_3) Oxides of nitrogen (NO, NO_2) Aldehydes (HCHO) Organic acids (HCOOH) Alcohols (CH_3OH) Hydrocarbons (CnHm) Carbon monoxides (CO) Hydrogen (H_2) Smoke

engines. First is reduction in the quantity of NOx formation during combustion of diesel fuel and second is reduction of NOx after formation i.e. diesel catalytic converter. Till now diesel catalytic converters are not in practice for reducing NOx. The three-way catalytic converter used in petrol driven vehicles cannot be used in the treatment of diesel engine exhaust gases specially NOx because the engine always operates with an overall lean air/fuel ratio and excess oxygen is present in the exhaust, though it can be used to oxidize the soot.

NOx reduction by Selective Catalytic Reduction (SCR) technique can occurs by four different gases, they are CO, H_2, NH_3 and HC. The reaction can be achieved efficiently in presence of these reducing gases in presence of excess oxygen.

Research is being carried out in many parts of the world to develop effective NOx reduction catalyst for use in diesel operated vehicles. But, no catalytic converter system has yet been commercialized that can eliminate nitrogen oxides (NOx) from the exhaust of vehicles powered by diesel and other lean-burn engines.

Origin of NOx

NOx is formed in the diesel engine by the reaction of atmospheric nitrogen and oxygen at high temperature. NOx accounts for 95% NO. From a thermodynamic point of view NO is unstable.

$$NO \rightarrow ½ N_2 + ½ O_2; ((\Delta G \pm 86 \text{ KJ/mol})$$

The NO is formed by the reaction between nitrogen & oxygen at high temperature by the following reaction mechanism [1].

$$N_2 + O^* \rightarrow NO + N^* \text{ ……………..(1)}$$

$$N^* + O_2 \rightarrow NO + O^* \text{ ……………..(2)}$$

$$N_2 + O_2 \rightarrow 2NO$$

By assuming a constant concentration of N* radicals and a large oxygen content when compared to NO concentration the following equation is obtained.

$$d[NO]/dt = 2k \exp(-EA/RT)[N_2][O^*] \text{ …………..(3)}$$

Equation (3) shows that formation of NO is controlled by the reaction (1). It is also revealed that the reaction temperature and atomic oxygen concentration are important parameters for NOx production and presence of nitrogen compounds in fuel oil is not significant towards NOx production [2].

NOx Effects on environment

NOx in the atmosphere is somewhat responsible for acid rain. The nitric oxide is rapidly oxidized by ozone while radicals such as OH* and HO_2 are transformed into NO_2, HNO_2 and HO_2NO_2 [3]. Acid rains usually formed at high altitude cloud and at that place where nitrogen oxides react with water, oxygen and other oxidants. These compounds are than transformed into nitric acid i.e. HNO_3 which acidifies the rain, snow and fog due to its solubility in water. The acid, when entering into the water bodies i.e. rivers, lakes and ponds affecting the eco-system and causing biological death of the water bodies.

NOx pollution is also promoting the formation of ozone, which is photochemical pollutant by the following reaction.

$$NO_2 + h\upsilon\ (\lambda < 415 \text{ nm}) \rightarrow NO + O^*$$

$$O_2 + O^* \rightarrow O_3$$

Ozone again reacts with NO and forms NO_2 and O_2.

$$O_3 + NO \rightarrow NO_2 + O_2$$

Formation of PAN i.e. peroxy acetyl nitrate (a photo-oxidizing pollutant responsible for smog formation especially during winter) is also due to presence of NOx in the atmosphere. PAN is produced by following chain reaction process [4].

$$NO_2 + h\upsilon \rightarrow NO + O^*$$

$$H_2O + O^* \rightarrow 2OH^*$$

$$CH_3 - CHO + OH^* \rightarrow CH_3 - CO^* H_2O$$

$$CH_3 - CO^* O_2 \rightarrow CH_3 - CO_3^*$$

$$CH_3 + CO_3^* NO_2 \rightarrow CH_3 - CO_3 - NO_2$$

It is clear that the NOx transformed pollutants HNO_3, O_3 and PAN are more harmful to human and plants.

NOx Effects on Human Health

Epidemiological studies have revealed that NOx concentration in the atmospheric air above 0.05 ppm for an exposure of above 24 hrs has hazardous effects on human health [5]. In air, NO is oxidized to NO_2.This process is rapid in high concentration of NO. In presence of sunlight and unburned hydrocarbon, this process is further accelerated. NOx is poisonous to human respiratory system, which provokes lung infection and respiratory allergies because it is able to diffuse through the alveolar cells and capillary vessels of the lungs and damage their structure through their propensity towards oxidation [6].

It should not be thought that the NOx has only hazardous effects, some new studies shows that a meager amount of NO is necessary for the living organism. Actually, NO is a neuro- messenger, which transmits the necessary information's to the WBC (white blood corpuscles) within the blood stream to destroy tumour cells and assorted bacteria. The next role is to the neurotransmitters to dilate the blood vessels. However, while the biologically active NO is a poisonous product of the in-vitro enzyme-catalyzed transformation of the amino acid into arginine. Therefore, higher concentration of NOx is toxic to human beings [7].

II. DIESEL EMISSION CONTROL TECHNOLOGIES

Diesel emissions can be controlled by modifying the in-cylinder combustion process or through exhaust after treatment. The primary concern is the emission of particulate and NOx. The various Diesel emission control techniques are as shown in Fig 7. The control methods can be divided by the type of diesel emission they can control and whether they are applied inside the cylinders of engine or after the exhaust gases emitted after combustion of diesel fuel in the engine [8].

Oxides of nitrogen can be controlled by modifying the peak temperature within the engine cylinder. The peak temperature can be reduced by charge air cooling i.e. reducing the air intake temperature into the cylinder or increasing the specific heat of the intake air i.e. exhaust gas recirculation or delaying the fuel injection time i.e. retarding the injection timing. These methods have been shown effective by some researchers [9, 10].

Charge air cooling and electronic timing control have already been introduced in present day engines. Further reduction in NOx can be anticipated by the use of NOx reduction catalyst i.e. Diesel catalytic converter especially for NOx [11].

The carbon particulate i.e. Soot portion can be controlled by improving the air-fuel mixing within the cylinder. Some typical methods are improving the atomization of fuel from the fuel injector by increasing the fuel injection pressure, forcing more air into the engine cylinder i.e. turbo-charging and improving the air/fuel mixing in the cylinder by modifying the combustion chamber. Application of these technologies to diesel engines has resulted in significant reduction in the soot portion of exhaust emission. Further reduction in soot particulate could be achieved by the use of soot filters.

The extractable portion of the particulate is controlled by reducing the sources of unburned fuel and lubricant from the engine. Some typical methods to reduce extractable particulate are improving the control of lubricant i.e. valve guide seals, improved cylinder kit and unburned fuel i.e. injector nozzle design. Improvement in engine design has resulted in a significant reduction in extractable particulate emissions. It is important to note that extractable particulate comprises of unburned fuel and lubricants. The high molecular weight hydrocarbons from the unburned lubricant represent a small portion of hydrocarbon emitted from the engine [12].

Thus, fuel derived hydrocarbon emission can reduce particulate emissions, but not completely. Further reduction could be achieved by the use of an oxidation catalyst in the catalytic converter system [13].

III. SELECTIVE CATALYTIC REDUCTION (SCR) TECHNIQUE FOR REDUCING EXHAUST EMISSION

i) **SCR by ammonia:** In this process (NH_3) ammonia is used as the reducing agent for nitrogen oxides to produce nitrogen and water [14].

$4NO + 4\,NH_3 \rightarrow 4N_2 + 6H_2O$

$6NO_2 + 8\,NH_3 \rightarrow 7N_2 + 12\,H_2O$

In these reactions presence of 0.1 -1% oxygen enhances the conversion of NOx and above 1% oxygen the conversion remains stable. An efficient catalyst for this process is V_2O_5 (vanadium pentoxide) supported over Al_2O_3, Al_2O_3 and SiO_2. This catalyst work good in the temperature range of 200^0C -550^0C and efficiency between 60-85%. Due to toxicity of ammonia, this technique is not used in motor vehicle but widely being used in thermal power plants worldwide.

ii) **SCR by hydrocarbons:** As discussed above, SCR by ammonia doesn't work for exhaust with excess oxygen, but when small amount of hydrocarbons are present in the motor exhaust, the catalyst works [15].

Active catalyst system for SCR by hydrocarbon can be divided into 3-groups namely Zeolites, Metallic oxides and noble metals.

(a) **Zeolites Group:** A study made by Iwamoto *et al* shows the effect of temperature upon the selective conversion of NO into N_2 when using the Cu ZSM- 5 catalyst. When the gas stream contains 1000 ppm NO, the catalyst is only 25% efficient at 673 K. The reaction can be completely inhibited by 1% oxygen in the reactant feed. However, when 166 ppm porpene is added in the stream, 100% reduction is obtained at 873 K [16]. Cu-exchanged zeolite, Pt-ZSM-5, Protonated zeolites, Zeolite containing gallium, cobalt and cerium some other varieties of Zeolite based catalyst systems.

(b) **Metal oxide Group:** Oxides such as Al_2O_3, SiO_2-Al_2O_3, TiO_2 and ZrO_2 are active for selective catalytic reduction of NO [17]. Alumina is most active with a reduction of 32% at 573 K (1000 ppm NO, 330 ppm C_2H_6, 10% O_2). Pure silica is not active. Supported transition metal oxide based SCR catalysts possess higher DeNOx activity in presence of hydrocarbons. Hamada *et al* have compared the activity of several metal oxides (Cu,Co,Ni,Mn,Fe) supported on Alumina or Silica. In general alumina supported catalysts are better than Silica supported catalysts. Miyadera [18] has studied activity of some transition metals (Cu, Co, Ag,V,Cr) supported on Alumina. Silver catalyst is most active giving 80% NOx conversion at 673 K. With these catalysts reducing agents like ethanol, acetone are most efficient reducing agents than propene. SCR by propane with oxides of Fe, Ag, V and Cr is also conducted by authors and it was found that Silver is most efficient SCR catalyst. The 5% silver catalyst has given maximum 43% and 5% iron catalyst has given maximum 39% NO conversion between 200-400^0C reaction temperature in the laboratory conditions [19].

(c) **Nobel metal Group**: Obuchi *et al* have studied SCR of NO by Platinum group of metals like Pt, Rh and Pd [20]. With alumina supported platinum the selectivity towards nitrogen is about 30% and other metals have shown nearly 50% nitrogen selectivity. But, platinum group metals don't work with excess oxygen in the exhaust gas stream.

IV. FUTURE FUELS FOR REDUCING EXHAUST EMISSION

In view of fast depleting fossil fuel reserve and to reduce the harmful exhaust emission gases emitted by burning the fossil fuels in automotive engines, some new and renewable fuels have been devised. These are Alcohols, Hydrogen, Biogas, Producer gas, LPG /LNG/ CNG, Synthetic fuels (i.e. GTL/CTL/BTL fuels), Methyl ester i.e. Bio-diesel (produced by esterification of edible & non edible oils), DME (Di-methyl ether) and ammonia.

For alternatives to diesel fuel, middle distillate fraction of GTL/ CTL/ BTL fuels is the new option. No SOx pollution is being created by these fuels and NOx pollution is less as compared to conventional diesel fuel. Methyl ester (Bio-diesel) produced by esterification of vegetable oils is now a day is mixed up to 20%. Many researchers have shown that there is a significant reduction in the exhaust emission by the use of Bio-diesel.

V. EXPERIMENTAL INVESTIGATION

The authors have conducted laboratory experiments [19] on DeNOx activity of four transition metal oxide (Fe, Ag, V and Cr) catalysts at 2, 5 and 8% metal loading over alumina support and in the stream of simulated exhaust gas. The catalytic activity has been tested between 100-500^{0}C reaction temperatures and GHSV (Gas hourly space velocity) between 6,000 h^{-1} to 18,000 h^{-1}. It was found that 5% silver catalyst has given maximum 43% and 5% iron catalyst has given maximum 39% NO conversion at 200-400^{0}C reaction temperatures.

On the basis of the above experiment, 5% Silver and 5% Iron catalysts were being prepared in bulk quantity of 430 gms (800 cc) over alumina support, photographs are shown in Fig-1 & 2. Two different design of catalytic converter was also fabricated for comparative study.

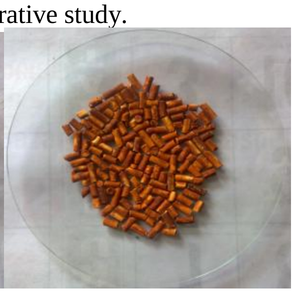

Fig.1 5% Silver Catalyst Fig.2 5% Iron Catalyst

These were tested in the exhaust gas stream of Jeep (Mahindra & Mahindra make) which is operated by diesel fuel. The exhaust gas velocity was measured by a vane type anemometer. During idling the exhaust gas velocity was 10.25 m/s and during full acceleration 32.0 m/s.

Two designs of "Diesel catalytic converter" were fabricated by 1.0 mm thick GI sheet. One is a Cylinder and another is Convergent-Divergent shapes which are shown in Fig: 3 and 4.

Fig.3 Cylindrical Catalytic Converter

Fig.4 Convergent-Divergent Catalytic Converter

The internal volumes of both the designs were kept same i.e. 800 cc. 5% Iron and 5% Silver loaded transition element catalyst (800 cc/ 430 gm) were prepared by wet impregnation technique and filled in these two catalytic converters and fitted in the exhaust pipe of Jeep. The catalytic converters were being heated externally by a 300 cm long 10 cm wide electrical heating tape by supplying external electricity. Field experiments are shown in Fig 5 & 6.

Fig.5 Experiment in Jeep Exhaust

Fig. 6 Exhaust Gas Analyzer

Temperature of the catalytic converter was raised slowly in steps from atmospheric temperature to 280^{0}C by a digital temperature indicator cum controller. Exhaust gas composition was measured by a Kane-May-UK make Flue Gas Analyzer. Table: 3, 4, 5 & 6 shows the experimental findings with 5% Silver & 5% Iron loaded catalysts which were filled in Cylindrical and Convergent-Divergent shape Catalytic converters.

VI. RESULTS AND DISCUSSION

When the Jeep exhaust gases passed through Cylindrical Catalytic converter fitted in the tail pipe of Jeep with 5% Silver loaded catalyst, the maximum NO conversion of 36.36% achieved at temperature of 280^0C under full acceleration. When experiment was conducted in Convergent-Divergent catalytic converter filled with 5% Silver catalyst the NO conversion slightly reduces to 31.41%. This is because the convergent-divergent shape gives less resistance in the flow of exhaust gases and consequently, the residence time is less for the reaction of nitric oxide gas with the catalyst.

Similarly, when 5% Iron catalyst was tested in Cylindrical catalytic converter, the NO conversion was 27.08% and with Convergent-Divergent catalytic converter it was 25.25% at 280^0C.

Graphical representations of the NO conversion efficiency of 5% Silver & 5% Iron catalysts with Cylindrical & Convergent-Divergent catalytic converter is shown in Fig 8.

VII. CONCLUSION

Transition metal oxide based catalytic converter containing 5% Silver and 5% Iron catalyst is an efficient system to reduce the NOx emission from diesel exhaust. Introduction of new future fuels will also be able to reduce NOx emissions in near future. The Cylindrical shape Catalytic converter offers the best shape for catalytic converter shell.

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TABLE 3 Exhaust components with 5% Silver catalyst in Cylindrical Catalytic converter

Gases	Concentration of exhaust gas components					
	At Idling & 50^0C	At full Acceleration & 50^0C	At full Acceleration & 100^0C	At full Acceleration & 150^0C	At full Acceleration & 200^0C	At full Acceleration & 280^0C
O_2 (%)	17.6	17.0	17.1	17.4	17.6	17.7
CO (PPM)	274	448	577	682	731	880
CO_2 (%)	1.9	2.2	2.1	2.0	2.0	1.9
NO (PPM)	128	143	134	126	115	91
NOx(PPM)	134	150	140	134	121	106

TABLE 4 Exhaust components with 5% Silver catalyst in Convergent-Divergent Catalytic converter

Gases	Concentration of exhaust gas components					
	At Idling & 50^0C	At full Acceleration & 50^0C	At full Acceleration & 100^0C	At full Acceleration & 150^0C	At full Acceleration & 200^0C	At full Acceleration & 280^0C
O_2 (%)	17.2	17.5	17.1	17.3	17.9	17.8
CO (PPM)	279	457	558	658	799	923
CO_2(%)	2.2	2.4	2.2	2.5	2.4	2.5
NO (PPM)	133	156	143	138	120	107
NOx(PPM)	140	163	151	144	127	114

Table 5 Exhaust components with 5% Iron catalyst in Cylindrical Catalytic converter

Gases	Concentration of exhaust gas components					
	At Idling & 50^0C	At full Acceleration & 50^0C	At full Acceleration & 100^0C	At full Acceleration & 150^0C	At full Acceleration & 200^0C	At full Acceleration & 280^0C
O_2 (%)	19.4	20.0	19.2	18.6	17.1	15.0
CO (PPM)	445	650	692	776	853	912
CO_2(%)	1.8	2.6	2.7	2.7	2.9	3.4
NO (PPM)	197	288	273	251	222	210
NOx(PPM)	205	295	281	259	230	216

Table 6 Exhaust components with 5% Iron catalyst in Convergent-Divergent Catalytic converter

Gases	Concentration of exhaust gas components					
	At Idling & 50^0C	At full Acceleration & 50^0C	At full Acceleration & 100^0C	At full Acceleration & 150^0C	At full Acceleration & 200^0C	At full Acceleration & 280^0C
O_2 (%)	17.7	18.8	18.1	17.9	17.4	17.2
CO (PPM)	460	678	697	781	884	933
CO_2(%)	2.6	2.8	2.7	2.9	3.3	3.3
NO (PPM)	201	297	275	267	231	222
NOx(PPM)	209	304	282	274	239	230

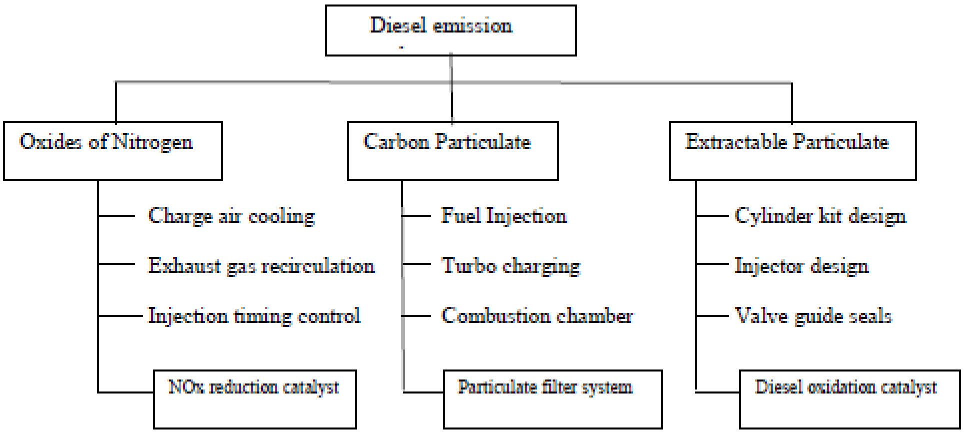

Fig. 7 Various Diesel emission control techniques

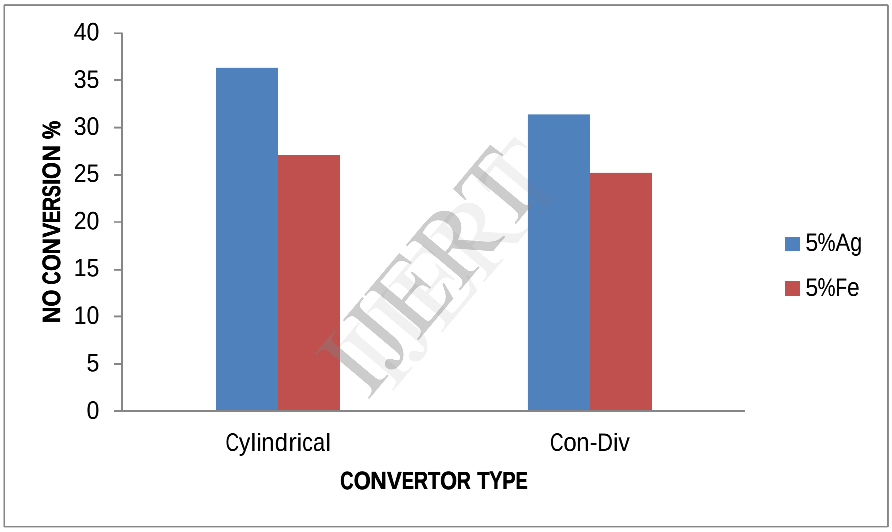

Fig.8 Comparison of Catalytic converter efficiency