Online NO_X Removal from Stationary Diesel Engine Exhaust by Barrier Discharge Plasma

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***Abstract* -- This paper reports plasma treatment of a 5 HP stationary diesel engine exhaust at full flow-rate (approximately 387 l/min) energized by power frequency AC. Glass and Chlorinated Polyvinyl Chloride (CPVC) tubes were used as the dielectric barriers. Significant removal of NO and NO_X has been achieved with barrier discharge plasma. The reactors were specially fabricated to facilitate high flow and high temperature associated with the engine exhaust. Studies were aimed at online cleansing of oxides of nitrogen from the exhaust without using any conventional catalysts/ adsorbents. The results were discussed in detail for the cases studied.**

***Index Terms*—NO_X removal, electric discharge plasma, online treatment, diesel engine exhaust**

I. Introduction

Air pollution is one of the major problems in urban areas both in developing and developed countries where many sources of airborne pollutants are concentrated. The main source of air pollutions is the combustion process of fossil fuels used in power plants, vehicles and other incineration processes. Due to the lower operating costs, diesel engines have been widely employed as the major propulsion source for heavy-duty transportations and off-road applications However, they emit NOx, CO, CO_2 and few other harmful gases leading to widespread pollution. Diesel engines alone account for nearly 48 percent of India's fuel consumption, thus contributing significantly to the man-made air pollution. One of the most harmful pollutants present in the diesel exhaust is NOx. The conventional techniques which are available to control emission now are either difficult to operate or does not satisfy the stringent emission standards. Reducing diesel engine exhaust to meet future emission standards is a challenging task and there is need for better after-treatment techniques [1].

For controlling NOx, the electric discharge based plasma is an upcoming technology being used mainly. Non-thermal plasma (NTP) technologies can be grouped as surface discharge, pulsed streamer corona discharge (PSC), dielectric barrier discharge (DBD) [2-4], and packed-bed corona discharge (PCD). These techniques have shown promising results owing to their rapid reactions, high electron energies and easy operation. There are many innovative non-thermal plasma techniques which have proved to be successful in controlling various air pollutants [5-14].

In this paper we describe a reactor configuration, where the full exhaust gas is being treated. This reactor configuration is used to treat the actual exhaust of a 5 HP diesel-generator set. Here power frequency AC has been used as the high voltage source for producing non-thermal plasma. Two type of reactor material are used, one with borosil glass and the other with chlorinated polyvinyl chloride (CPVC). The main emphasis is laid on the NO_X treatment of diesel engine exhaust. Experiments were carried at two different engine loadings.

II. Experimental Setup

The schematic of experimental setup used during the present research work is shown in Fig. 1. Studies were carried out on full exhaust of a diesel generator set and main emphasis has been given on removal of NOx. The raw diesel exhaust gas is allowed to pass through the reactor for treatment. For the analysis of pollutants a sample gas is taken through particulate filter (for the protection of flue gas analyser) to the analyser.

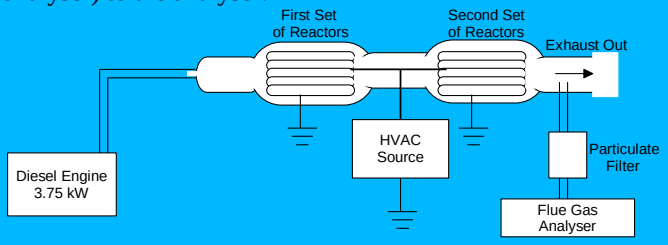


Fig. 1. Line diagram of experimental setup.

A. *Diesel Engine*

The diesel exhaust for the treatment was taken from a 5HP (3.75kW) diesel generator. The loading of diesel engine is done electrically. Depending on the atmospheric conditions the concentrations of the exhaust varies which was closely monitored during the course of experiments. The diesel engine is operated at no load and 20% load. The entire exhaust is allowed to pass through the reactor set during the experiment. The gas flow rate is about 387 l/min.

B. *High Voltage Source and Measuring System*

The power source consists of a 230 V/50 kV step-up transformer. Voltage parameters were measured by using a voltage divider of ratio 2000:1 (EP-50 K, PEEC) and the

digital storage oscilloscope (DL 1540: 8 bits 200 MS/s, 150 MHz, Yokogawa). The consumed power was measured from the input side by two reactor principles using a digital wattmeter [15]. Fig. 2 shows the power input to the reactor at different input voltage. It has been observed that with the increase in engine load the exhaust temperature has increased. The exhaust temperature is found to be 55 °C and 120 °C at no load and 20% load respectively. The power input to the reactor increases as the engine loading is changed from no load to 20% load. This input power can be expressed as specific energy density (SED) in J/L as follows:

$$\text{Specific Energy Density (SED)}: \frac{\text{Power input to the reactor (watt)}}{\text{Gas flow rate (l/sec)}} \quad (2)$$

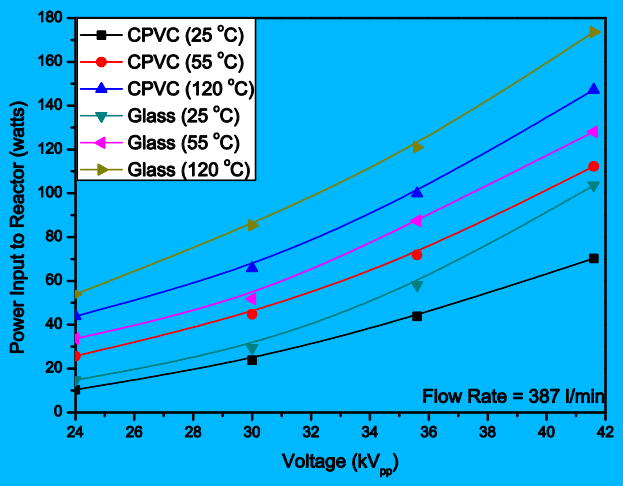
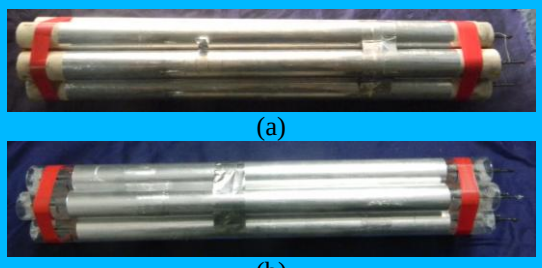


Fig. 2. Power input to reactor set at different voltage

C. Plasma Reactor

The plasma reactor is one in which the high voltage is applied to produce the much needed electrical discharge to initiate the reaction to remove/convert NO and NOx emissions from the diesel exhaust.



(a)

(b)

Fig. 3. Plasma reactor (a) CPVC (b) glass

In general, the plasma reactor consists of a stainless steel electrode placed inside a glass tube/CPVC. Each of the reactor set consists of seven tubular reactor bunched together. Fig. 3 shows the picture of plasma reactors used during the experiment. The outer and inner diameter of CPVC tube used is 28 mm and 25 mm respectively, and that of glass is 30 mm and 27 mm. The electrode used is of stainless steel, which has a diameter of 2 mm.

D. Flue Gas Analyser

The gas after the treatment in the plasma reactor, a sample gas is let into the flue gas analyzer to analyze the proportions of the gas components. For each engine load condition the concentration of the exhaust is recorded. The analyzer records the concentration of various gases like NO_x, NO, NO_2 CO, CO_2, and O_2 by a set of electro chemical sensors. The analyzer has a pump that is capable of taking the exhaust gas at 2 l/min from the exhaust pipe line.

III. Results and Discussion

Here, we discuss the results of the experiments carried out on a stationary diesel engine exhaust using the setup described in Fig. 1. The experiments have been conducted on entire gas stream at no load and 20% load conditions. Power frequency AC voltage was applied in the range of 24-42 kVpp. The input power to the reactor was measured by using two reactor methods and then expressed as specific energy density (SED) in J/L. The initial concentration of pollutants in diesel engine exhaust is given in Table I. Though different types of hydrocarbons like toluene, methanol, phenol, aldehydes etc., are also present in the diesel exhaust, they could not be measured due to the limitations of the gas analyzer used.

TABLE I
Initial Concentration of Diesel Engine Exhaust

POLLUTANTS	*DIESEL ENGINE AT No Load*	*DIESEL ENGINE AT 20% Load*
NO	85	234
NO_2	44	53
NO_x	129	287
CO	919	1339
CO_2	0.1 %	0.1 %
O_2	17.7 %	15.7 %

A. Removal of NO/NOx in diesel engine exhaust using CPVC tube

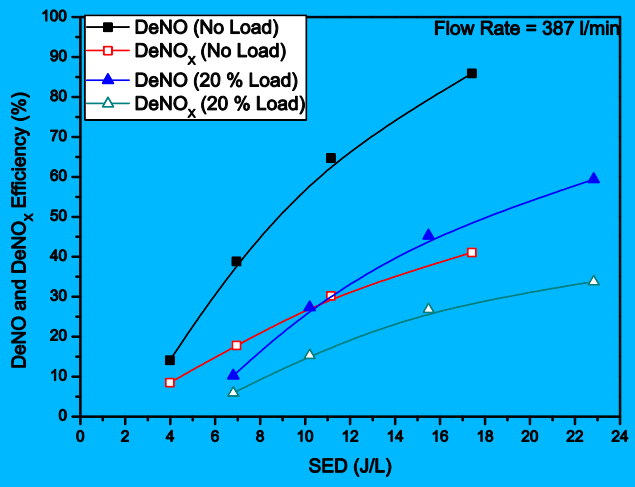


Fig. 4. DeNO and $DeNO_X$ efficiency with one CPVC reactor set at different load

Studies were conducted with entire stream of raw diesel exhaust gas. First the performance for NO and NO_X removal was evaluated with one set of rector consisting of CPVC tubes, then studies were carried out with two such reactor set in cascade.

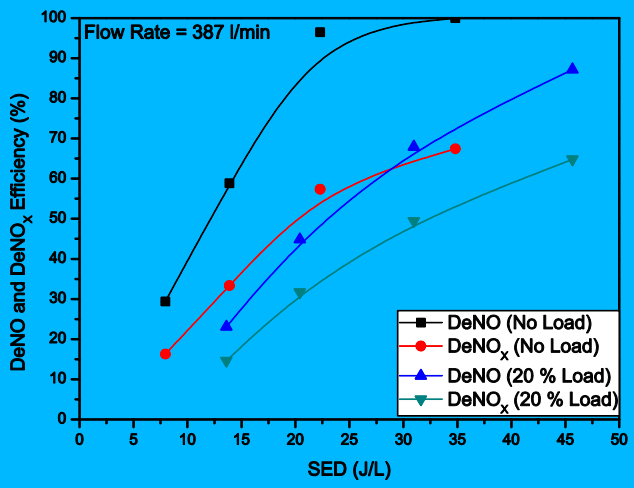


Fig. 5. DeNO and $DeNO_X$ efficiency with two CPVC reactors set in cascade at different load

Fig. 4 shows the NO and NOx removal efficiency with one CPVC reactor set being energized. At no load with the full exhaust the DeNO and DeNOx efficiency is 85% and 41% respectively with an energy input of 17 J/L. When the load is increased to 20% the DeNO efficiency is reduced to 59% with an increased SED of 22 J/L. At this energy input the NOx removal is found to be 33%. This may be due to the increased initial concentration of NOx from no load to 20% load. Studies were also carried out with two reactor set in cascade.

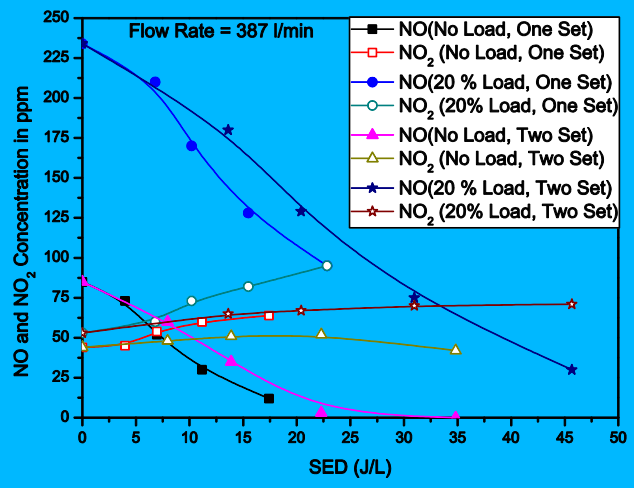


Fig. 6. Variation of NO and NO_2 concentration with SED using one reactor set and two CPVC reactors set in cascade at different load

From Fig. 5, it is observed that with cascaded reactors set 100% NO conversion has been achieved with 34 J/L at no load, whereas the NOx removal is about 67%. When, the engine is operated at 20% loading, the NO conversion was about 87% with an energy input of 45 J/L. At this energy input the cascaded reactor set shows a maximum NOx removal of 64%. It has been observed that with cascading two reactors set the energy input gets doubled with significant increase in efficiency of NOx.

Here it has been observed that the $DeNO_X$ efficiency is mainly due to the reduction /conversion of NO. In case of diesel engine exhaust the conversion NO to NO_2 takes place due to oxygen as well as ozone. It may be inferred from (2, 4) that, NO conversion from ozone radical induced reaction is more dominant than oxygen radical induced reaction. Further, the NO_2 so formed gets converted to NO_3 or N_2O_5 or back to NO. The latter two reactions are more or less equally probable and the former one i.e. NO_3 formation appears to be more dominant than the other two. It is clear that high electric field gives higher energetic electrons. These high energetic electrons collide with other background gas molecules, resulting in the production of more excited species and ions. However, most of them get quickly converted to radicals [1]. Possible reaction pathways responsible for NO and NOx removal can be summarized as below [11]:

NO-NO_2 conversion reactions involving $O/O_3/NO_3$ radicals

$$O_2 + e \rightarrow O + O + e \qquad k = 1.1\times10^{-12}\ cm^3/s \qquad (1)$$

$$NO + O \rightarrow NO_2 \qquad k = 1.4\times10^{-12}\ cm^3/s \qquad (2)$$

$$O_2 + O \rightarrow O_3 \qquad k = 1\times10^{-14}\ cm^3/s \qquad (3)$$

$$NO + O_3 \rightarrow NO_2 + O_2 \qquad k = 1.8\times10^{-14}\ cm^3/s \qquad (4)$$

$$NO + NO_3 \rightarrow NO_2 + NO_2 \qquad k = 1.5\times10^{-11}\ cm^3/s \qquad (5)$$

NO reduction reactions

$$N_2 + e \rightarrow N + N + e \qquad k = 3.0\times10^{-16} \qquad (6)$$

$$NO + N \rightarrow N_2 + O \qquad k = 2.1\times10^{-11} \qquad (7)$$

NO_2 conversion reaction

$$NO_2 + O \rightarrow NO + O_2 \qquad k = 10^{-11}\ cm^3/s \qquad (8)$$

$$NO_2 + O_3 \rightarrow NO_3 + O_2 \qquad k = 1.8\times10^{-17}\ cm^3/s \qquad (9)$$

$$NO_2 + NO_3 \rightarrow N_2O_5 \qquad k = 1.8\times10^{-11}\ cm^3/s \qquad (10)$$

$$NO_2 + N \rightarrow N_2O + O \qquad k = 3\times10^{-12}\ cm^3/s \qquad (11)$$

(Where k = rate constant)

Further, the partly converted NO_2 also reacts with soot [2] by the following reaction

$$NO_2 + Soot \rightarrow CO + CO_2 + N_2 + H_2O + NO \qquad (12)$$

Diesel exhaust being an oxygen rich exhaust contains large number of O radicals, which enables the efficient oxidation of NO to NO_2. This hypothesis is in good agreement with the studies by other researchers [16]. The variation of NO and NO_2 concentration in ppm is shown in Fig. 6. The initial concentration of NO and NO_2 were 85 ppm and 44 ppm at no load respectively which got increased to 234 ppm and 53 ppm at 20% load. Due to the strong oxidation property of plasma well known decrement of NO and increment of NO_2 occur. It has been observed that the increment of NO_2 in case of two reactor set is not much; the initial 53 ppm of NO_2 increased to 71 ppm only at 20% load whereas with one reactor set the initial 53 ppm increased to 95 ppm.

B. Removal of NO/NOx in diesel engine exhaust using glass tube

First the performance for NO and NO_X removal from a diesel engine exhaust was evaluated with one set of rector consisting of glass tube then studies were carried out with two such reactor set connected in cascade.

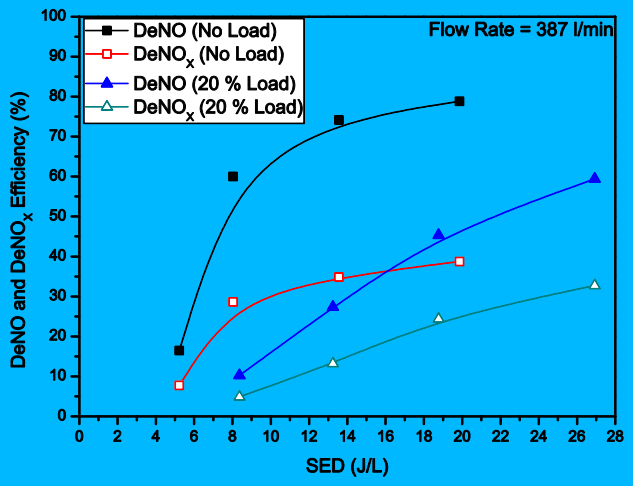


Fig. 7. DeNO and $DeNO_X$ efficiency with one glass reactor set at different load

It has been observed here in case of glass tube reactor the power drawn from source is more compared to glass tube. But the DeNO and DeNOx efficiency is more or less same with an increase in SED value. Fig. 7 shows the NO and NOx removal efficiency with one glass reactor set being energized. DeNO and DeNOx efficiency found to be 78% and 38% respectively with an energy input of 20 J/L at no load while treating entire exhaust stream. When the load is increased to 20% the DeNO efficiency is reduced to 59% with an increased SED of 27 J/L. At this energy input the NOx removal is found to be 32%. This may be due to the increased initial concentration of NOx from no load to 20% load.

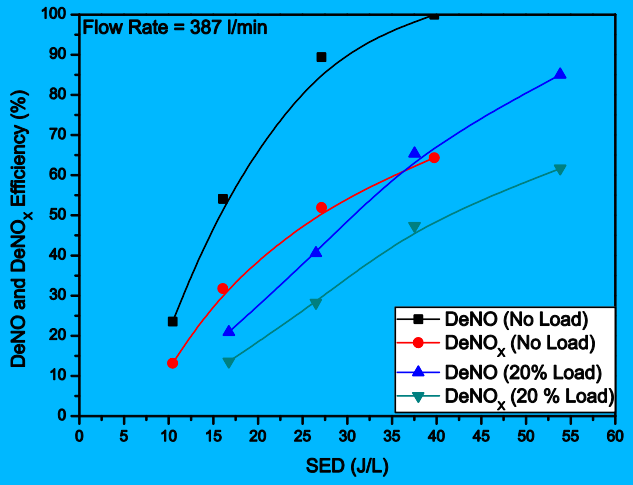


Fig. 8. DeNO and $DeNO_X$ efficiency with two glass reactors set at different load

Studies are also carried out with two reactors set in cascade. From Fig. 8, it is observed that with cascaded reactors set with an energy consumption of 39 J/L, the NO conversion is 100%, whereas the NOx removal is about 64%. When, the engine is operated at 20% loading, the NO conversion came down to 85% with an energy input of 53 J/L. At this energy input the cascaded reactor set shown a maximum NOx removal of 61%.

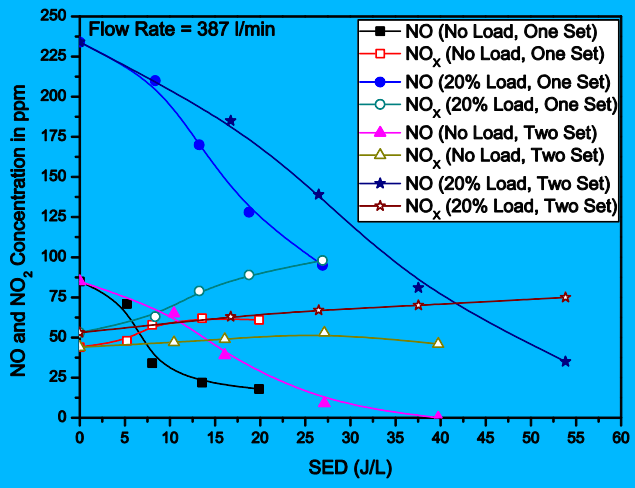


Fig. 9. Variation of NO and NO_2 concentration with SED using one reactor set and two glass reactors set in cascade at different load

The variation of NO and NO_2 concentration in ppm is shown in Fig. 9. The initial concentration of NO and NO_2 were 85 ppm and 44 ppm at no load respectively which got increased to 234 ppm and 53 ppm at 20% load. The initial 53 ppm of NO_2 increased to 71 ppm only at 20% load whereas with one reactor set the initial 53 ppm increased to 95 ppm. At no load with two reactor set in cascade the NO concentration goes to zero value from 85 ppm and the NO_2 value initially increased to 53 ppm at 27 J/L whereas, further increase in SED decreases the NO_2. This might be due to the fact that at 27 J/L the NO concentration has almost diminished.

IV. Conclusions

A reactor has been designed, built and tested with full flow of diesel engine exhaust. This reactor can be used for large amount of exhaust gas without significant design modification. The reactor can be easily scaled to the required size for removal of pollutants. The discharge plasma exhibits reasonable performance in exhaust treatment. For enhancing the removal capability two cascade reactor systems have been used. Among two type of reactor observed the CPVC reactor was energy efficient compared to glass reactor with the application of HVAC.

It has been seen from the results, the removal efficiency decreases as the engine load increases. But it has been observed that the decrease in efficiency is not large. NO_X removal of 64% was achieved at a flow rate of 387 l/min with CPVC reactor at an energy input of 45 J/L. Whereas, for achieving $DeNO_X$ efficiency of 61% at flow rate of 387 l/min

the glass reactor consumed 53 J/L. The CPVC reactor is energy efficient compared to glass reactor when operated with power frequency AC at higher gas flow rate. The studies indicate that there is a possibility of retrofitting the stationary diesel-generator set with non-thermal plasma reactor for significant control of NO/NOx instead of resorting to conventional adsorbent based removal techniques. Further, the commercially available HVAC sets can be used for the NTP generation in combination with CPVC reactor tubes.

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