Study of Pulsed Plasma in a Crossed Flow Dielectric Barrier Discharge Reactor for Improvement of NO_x Removal in Raw Diesel Engine Exhaust

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Abstract Improved performance of plasma in raw engine exhaust treatment is reported. A new type of reactor referred to as of cross-flow dielectric barrier discharge (DBD) was used, in which the gas flow is perpendicular to the corona electrode. In raw exhaust environment, the cross-flow (radial-flow) reactor exhibits a superior performance with regard to NO_X removal when compared to that with axial flow of gas. Experiments were conducted at different flow rates ranging from 2 L/min to 25 L/min. The plasma assisted barrier discharge reactor has shown encouraging results in NO_x removal at high flow rates.

Keywords: electric discharges, NO_x removal, non-thermal plasma, raw diesel engine exhaust

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1 Introduction

Diesel engines, one of the major power sources in industries and automobiles, play a significant part among man-made pollution sources. Diesel today accounts for nearly 48% of India's fuel consumption while the share of petrol is below 7%. Thus control of diesel engine exhaust is important. Successful control of emissions from combustion engines particularly from diesel engine is yet to be achieved. Now the conventional techniques available to control emission are either difficult to operate or do not satisfy the stringent emission standards. Amongst the gaseous pollutants in diesel exhaust, the major concern and a challenging task is to control oxides of nitrogen, commonly referred as NO_x. In the case of diesel engines, despite the modification in engine design and improvement in post-treating technologies, large amount of NO_x continuous to get emitted and attempts to develop a new catalyst to reduce NO_x, have been less successful yet and appear to be more expensive. Furthermore, with the increasingly stringent emission standard, estimates are that NO_X must be reduced by as much as 90% from engine-out levels by 2010. It is in this context the use of plasma for the treatment of gaseous pollutants in the diesel exhaust has emerged as a promising alternative, at least, at the laboratory levels [1~6].

The basic challenge with this technique lies in reducing the power consumption of the pulsed corona device while treating higher flow rates of gas. Pulse corona reactor is simple and widely used. Although it is simple, this geometry is impractical for the case with high flow rate. However, when reactors are stacked in parallel there is a possibility of treating larger volumes of gas [7]. The power consumption with small gas flow rate has already be reduced to 2 L/min to 5 L/min. However no much work is reported in literature with high gas flow rates treated with non-thermal plasma [8~10]. An experimental study of DC and pulsed corona discharge for NO_x reduction from diesel engine exhaust at very high flow rates has been reported and results have been expressed in terms of cleanness and efficiency of NO_x removal [11,12].

In this paper an alternative reactor geometry referred to as cross-flow DBD reactor is described, where the exhaust gas flows perpendicular to the wire-cylinder reaction chamber. This reactor is used to treat the actual exhaust of a 3.75 kW diesel-generator set. The main emphasis is laid on the NO_x treatment of diesel engine exhaust.

2 Experimental setup

The schematic of experimental setup used during the present research work is shown in Fig. 1. Studies were carried out at the laboratory scale using a diesel generator set as the source of the exhaust and main emphasis was given on removal of NO_x. Sample gas from the diesel engine exhaust pipe is first allowed to pass through steel wool where the coarse oil mist and dust particulate get trapped. The raw sample is then allowed to flow through the reactor for treatment. Proper care was taken to protect the gas analyzer from dust contamination.

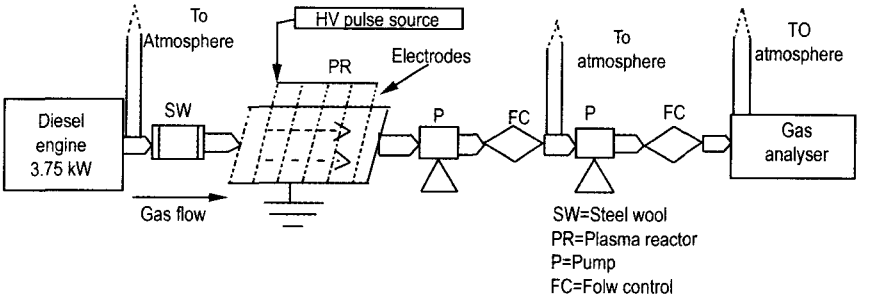


Fig.1 Schematic diagram of experimental setup used for the experiment

2.1 Exhaust source

For the real exhaust, diesel engine (Make: Sardhara Engine manufacturers, Rajkot, India, air cooled, 5 HP, 3.7 kW, 1500 RPM, year of production: 2009) was used as a source for gaseous pollutants. A part of the exhaust gas was made to pass through the plasma reactor. The gas flow rate in the treatment zone varied from 2 L/min to 25 L/min. The loading of diesel engine is carried out electrically. Depending on the atmospheric conditions the concentrations of the exhaust varies which was closely monitored during the course of experiments.

2.2 Pulse generation using rotary spark gap

This set up consists of a single-phase AC supply, a rectifier and a capacitor. Fig. 2 shows the basic circuit diagram for generating high voltage pulse using rotary spark gap. The capacitor C_g is charged to a DC voltage through a rectifier. When the rotating electrode of RSG (rotary spark gap) is aligned with the fixed line electrode, the charge on C_g is discharged to the load (plasma reactor). The hemispherical rotating electrode of the RSG is connected to a motor through an insulating shaft. The frequency of the pulses applied to the reactor is controlled by changing the speed of the motor. In the present study, the pulse rate was kept constant at about 75 pulses per sec. The rising time of the pulses used was around 20 ns. The pulse voltage generated from the rotary spark gap is shown in Fig. 3. The pulse is approximately of square one with a duration of 13 ms.

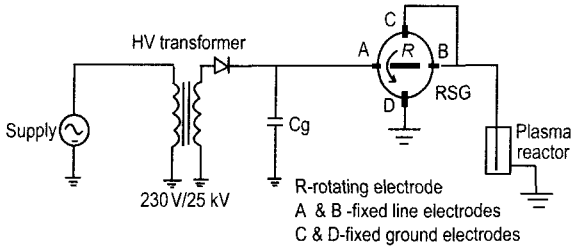


Fig.2 Sketch of high-voltage repetitive pulse generator

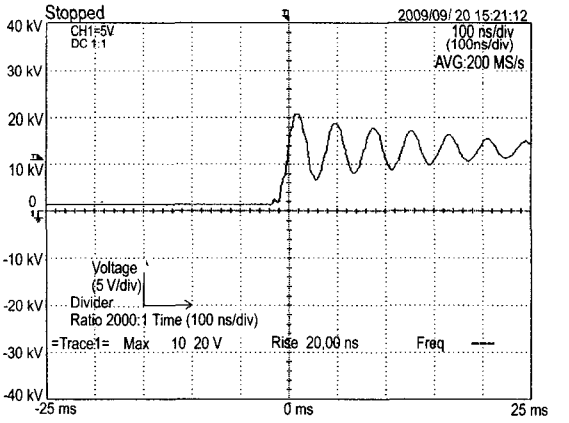


Fig.3 Rising portion of pulse voltage waveform across the reactor

2.3 Measuring system

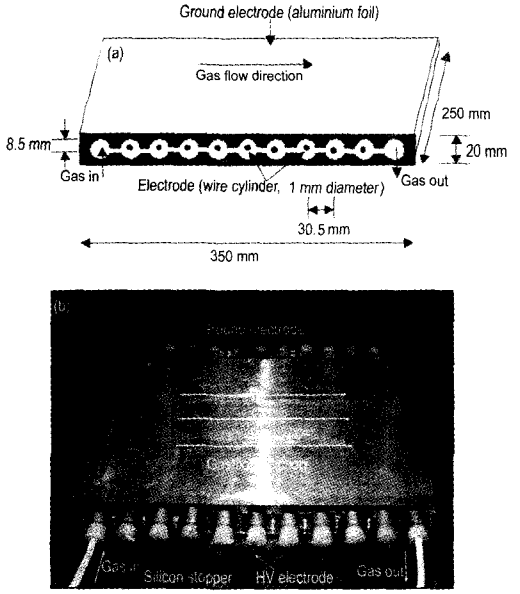
The measurement of the concentration of NO_X and the other soecies is carried out by the combustion analyzer (Quintox KM 9106). This combustion analyzer uses electro-chemical principle for the analysis of different gases in the diesel exhaust. Pulse parameters were measured by using a voltage divider of ratio 2000 : 1 (EP-50 K) and the digital oscilloscope (DL 1540: 8 bits 200 MS/s, 150 MHz, Yokogawa). The consumed power was measured from the input side by two-watt meter principle using a digital wattmeter. Further specific energy density (SED) can be calculated from the following formula:

$$\text{Specific Energy Density (SED) (J/L)} = \frac{\text{Power input to Reactir(W)}}{\text{Gas Flow Rate (L/s)}}.$$

2.4 Gas treatment reactor

The reactor is a rectangular cuboid made up of acrylic (350 mm × 250 mm × 20 mm) and is named as cross flow dielectric barrier discharge reactor. Fig. 4(a) and (b) shows the schematic and actual reactor, respectively. The reactor is wrapped uniformly with an aluminum foil and precaution was taken while wrapping such that no air gap should be present as it leads to local

breakdown and hence, power loss. The aluminum foil was grounded, which acts as grounded electrode. For inner corona electrode stainless steel material is chosen to avoid corrosion. The cuboid consists of eleven number of tubular shape with outer diameter of 20 mm and inner diameter of 17 mm. Provision has been made such that gas will flow radially instead of axially. Out of eleven number of reactor, only nine were energized with high voltage pulse. The effective corona region is about 210 mm.



(a) Schematic, (b) Real

Fig.4 Cross flow DBD reactor

3 Results and discussion

The experimental setup is described schematically in Fig. 1. Cross flow DBD reactor was used to treat the exhaust gas. A new type of reactor was designed in which gas was flowing radially instead of axially. The high voltage source is used with pulse of repetitive rate 75 cycles per second and rising time of 20 ns. The exhaust composition of simulated gas and diesel engine at load is listed in Table 1. The experiments were conducted for different flow rates starting from 2 L/min, 5 L/min, 10 L/min, 15 L/min, 20 L/min and 25 L/min.

Table 1. Initial concentration of diesel engine exhaust

Pollutants	Diesel exhaust at 27% load
NO	350 ppm
NO_2	50 ppm
NO_x	354 ppm
CO	2500 ppm
CO_2	0.3% vol
O_2	15.4% vol

3.1 DeNO/DeNO$_X$ experiment on diesel engine exhaust

Experiments were conducted using a diesel generator set of 3.75 kW under a loaded condition of 27%. Table 1 lists the initial concentration of the pollutants present in diesel engine exhaust. The concentration of NO and NO_2 were measured individually and then added to get the NO_x concentration. 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.

Studies were conducted with raw diesel engine exhaust gas and it was shown that the use of plasma leads to an improved performance while treating raw diesel engine exhaust [13]. Thereby it is suggested to apply for practical situation. First the performance of the cross flow DBD reactor for NO and NO_x removal from a diesel engine exhaust was evaluated with flow rate maintained at 2 L/min. The DeNO and DeNO$_x$ efficiency is shown in Figs. 5 and 6, respectively. Here it is observed that the NO removal efficiency in the case of nine reactors is 100% at a specific energy density of 486 J/L, whereas it is 74% at 54 J/L for one reactor. For NO_x removal the same phenomenon was observed, i.e., the NO_x removal efficiency is almost 50% higher for nine reactors than one reactor. From Figs. 5 and 6

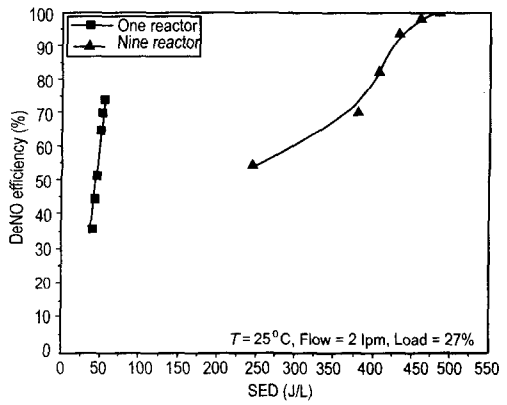


Fig.5 NO removal efficiency as a function of SED (J/L) value for different flow rates

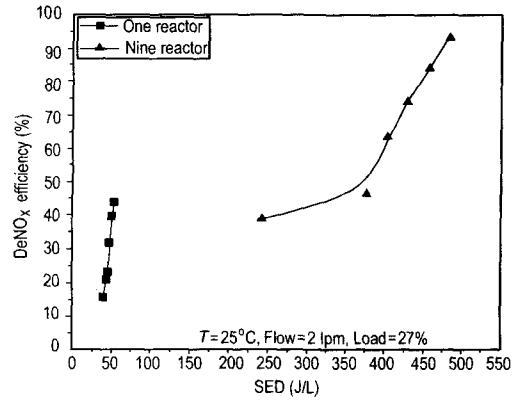


Fig.6 NO_x removal efficiency as a function of SED (J/L) value for different flow rates

it is found that the removal efficiency is much higher for nine reactors rather than for one reactor. Further experiments are carried out with nine reactors energized for different flow rates ranging from 5 L/min to 25 L/min.

The specific energy density does not vary with the increase in gas flow rate [8]. With high O_2 concentration, large number of O radicals will be formed which enables efficient oxidation of NO to NO_2. In Fig. 7 it is seen the variation of NO and NO_2 concentration in ppm for gas flow rate of 15 L/min and 20 L/min. The initial concentration of NO and NO_2 were 233 ppm and 62 ppm, respectively, when the gas flow rate was 15 L/min. As the input energy density increases the NO gets oxidized to NO_2 thus the concentration of NO decreases while that of NO_2 increases. Finally at SED value of 65 J/L the concentration of NO reached to 36 ppm and NO_2 to 98 ppm. The same oxidation occurs for the entire flow rate. The concentrations of NO and NO_2 with their initial values of 256 ppm and 54 ppm changed to 63 ppm and 98 ppm, respectively, at a SED of 49 J/L with a gas flow rate of 20 L/min.

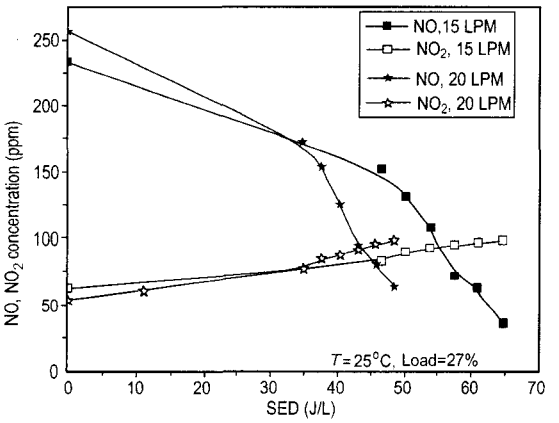


Fig.7 NO and NO_2 concentration as a function of SED value

Here for diesel engine exhaust the removal of NO takes place mostly by the conversion of NO to NO_2 due to oxidation of O/OH radicals. It is clear that high electric field leads to highly energetic electrons. These highly energetic electrons collide with other background gas molecules, resulting in the generation of more excited species and ions. However, most of them get quickly converted to radicals [14]. Possible reaction pathways responsible for NO and NO_x removal can be summarized as follows [15]:

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

$$O_2 + e \rightarrow O + O + e \quad (1)$$

$$NO + O \rightarrow NO_2 \quad (2)$$

$$O_2 + O \rightarrow O_3 \quad (3)$$

$$NO + NO_3 \rightarrow NO_2 + O_2 \quad (4)$$

$$NO + NO_3 \rightarrow NO_2 + NO_2 \quad (5)$$

NO reduction reactions

$$N_2 + e \rightarrow N + N + e \quad (6)$$

$$NO + N \rightarrow N_2 + O \quad (7)$$

NO_2 conversion reaction

$$NO_2 + O \rightarrow NO + O_2 \quad (8)$$

$$NO_2 + O_3 \rightarrow NO_3 + O_2 \quad (9)$$

$$NO_2 + O_3 \rightarrow N_2O_5 \quad (10)$$

$$NO_2 + N \rightarrow N_2O + O \quad (11)$$

Furthermore, the partly converted NO_2 also reacts through the following reaction [16]:

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

In Fig. 8 the removal efficiency of NO against specific energy density, for different gas flow rates, is shown. It is seen that NO removal is higher at a gas flow rate of 5 L/min. The NO removal efficiency is 48% at 140 J/L which increases to 100% at 194 J/L with a gas flow rate of 5 L/min. This removal efficiency decreases for a higher flow rate. But still at a higher flow rate, e.g., 25 L/min, the NO removal was quite significant. For a further increase in the flow rate the required specific energy density also gets reduced. A 68% removal of NO can be seen in Fig. 8 with a gas flow rate of 25 L/min at SED of 39 J/L.

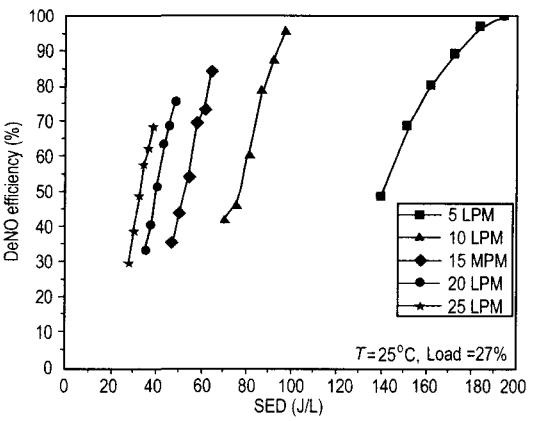


Fig.8 NO removal efficiency as a function of SED (J/L) value for different flow rates

The removal efficiency is quite high in the case of nine reactors because the particulate stay longer duration across corona discharge. The NO_X removal efficiency against energy density can be observed in Fig. 9. As in the case of NO removal efficiency the efficiency was also high for a low flow rate, and decreases further when the gas flow increases. The NO_X removal efficiency was around 44% at an input energy density of 39 J/L.

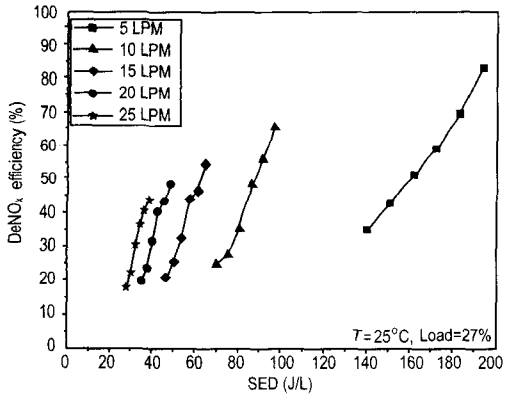


Fig.9 NO_x removal efficiency as a function of SED (J/L) value for different flow rates

From an industrial point of view it is very much important to know the energy needed for NO and NO_x molecule to be removed. The eV/NO and eV/NO_x can be calculated using the following formula:

Energy cost Pet NO molecule=

$$\frac{\text{power input to reactor } \times 6.25 \times 10^{18} \text{ eV/sec}}{\text{NO molecule remove}},$$

where NO molecules removed $= \eta_{\text{DeNO}} \times \text{NO}_{\text{Initial}}(\text{ppm}) \times 10^{-6} \times 2.45 \times 10^{19}$ (molecules/cc)$\times$ gas flow (cc/sec).

Energy cost per NO_x molecule=

$$\frac{\text{power input to reactor} \times 6.25 \times 10^{18} \text{ eV/sec}}{\text{NO}_x \text{ molecule removed}},$$

and

NO_X molecules removed $= \eta_{\text{DeNO}_x} \times \text{NO}_{x\ \text{Initial}}(\text{ppm}) \times 10^{-6} \times 2.45 \times 10^{19}$ (molecules/cc)$\times$ gas flow (cc/sec).

The required eV/NO molecule as a function of NO removal efficiency is shown in Fig. 10. It is seen that at lower removal efficiency the energy in eV required for NO molecule removal is quite high. As the removal efficiency increases the eV/NO molecule get reduced for a fixed gas flow rate. Furthermore, eV/NO molecule reduced significantly with the increase in flow rates. At a gas flow of 5 L/min and lower value of SED the removal efficiency of NO is low which in turn the eV/NO molecule gets high, about 385 eV. As the SED increase

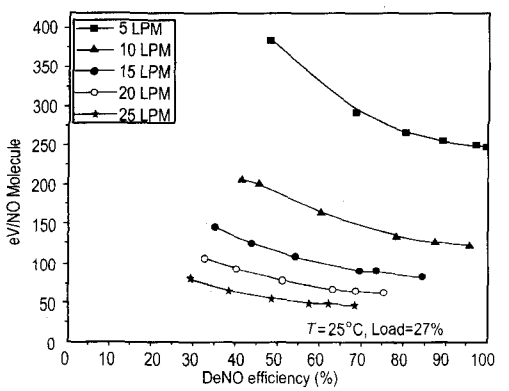


Fig.10 eV/NO molecule as a function of DeNO efficiency for different flow rates

further to 194 J/L the eV/NO molecule falls to 258 eV. This happens because eV/NO molecule depends on the NO removal efficiency. The eV/NO molecule further decreases when the gas flow increases. At a gas flow rate of 25 L/min only 47 eV is required per NO molecule being removed at SED value of 39 J/L.

In Fig. 11 the eV/NO_x molecule removed with respect to $DeNO_X$ efficiency is shown. It is seen that for a NO_X removal efficiency of 44% and a gas glow rate of 25 L/min only 63 eV/NO_X molecule was needed, whereas at a gas flow rate of 5 L/min, eV/NO_X molecule increases to 378 eV.

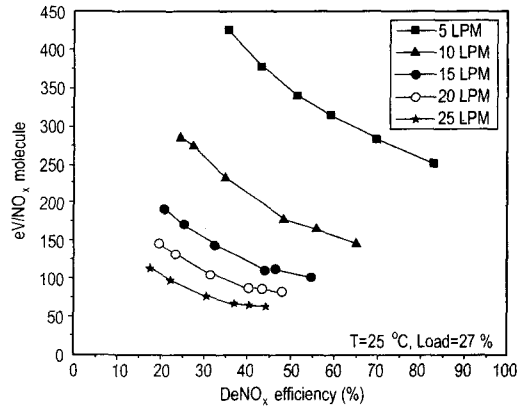


Fig.11 eV/NO_x molecule as a function of $DeNO_x$ efficiency for different flow rates

The removal of NO and NO_X in terms of gm/kWh w.r.t. input specific energy density is shown in Figs. 12 and 13, respectively. In Fig. 12, the NO removal for 5 L/min case is 4.44 gm/kWh at an input energy density of 194 J/L. But for a gas flow rate of 25 L/min the NO removal is 24 gm/kWh at a SED of 39 J/L.

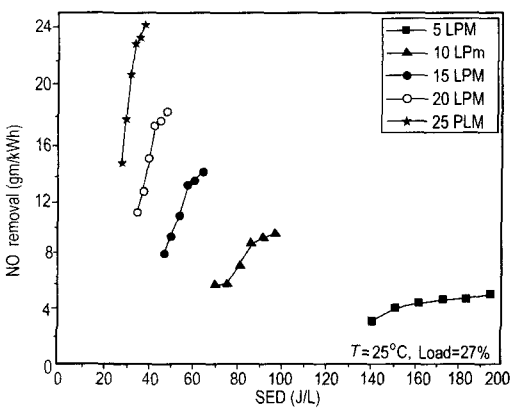


Fig.12 NO removal in gm/kwh as a function of SED (J/L) value for different flow rates

The NO_X removal in terms of gm/kWh w.r.t. SED (J/L) is shown in Fig. 13. NO_X with 18 gm/kWh was removed from raw diesel exhaust for the gas flow rate restricted to 25 L/min. The input specific energy density requirement was only 39 J/L.

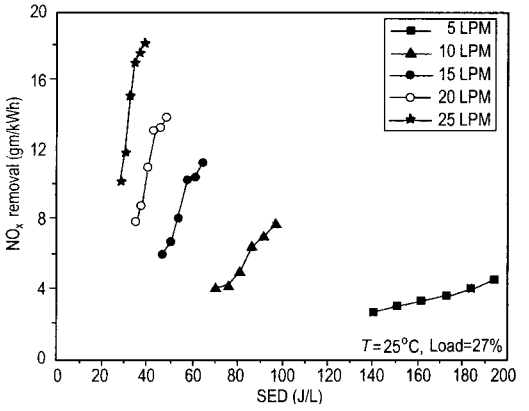


Fig.13 NO_x removal in gm/kWh as a function of SED (J/L) value for different flow rates

A summary of $DeNO_x$ efficiency for the entire flow rate considered in terms of cfm (1 cfm = 28.32 L/min) for different SED (J/L) values is shown in Fig. 14.

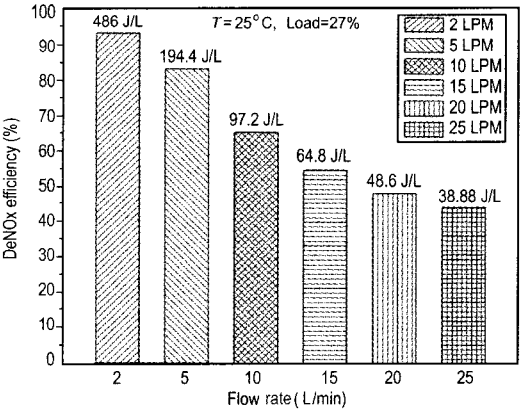


Fig.14 $DeNO_x$ efficiency as a function of flow rates for different specific energy density

4 Conclusion

A new cross flow DBD reactor was designed, built up and tested with diesel engine exhaust. This reactor can be applied to large amount of exhaust gas without significant design modification. This reactor can be easily scaled to the required size for removal of pollutants. The plasma exhibits an improved performance in raw diesel engine exhaust treatment, which is attributed to the presence of soot and oil mist in the raw exhaust.

It was seen from the results, the removal efficiency decreases as the gas flow rate increases. But it was observed that the decrease in efficiency is not large. NO_x removal as high as 45% was achieved at a high flow rate of 25 L/min. The energy consumed was 64 eV/NO_x (18 gm/kWh). The result was achieved without using any catalysts/adsorbents. Thus with less amount of energy cross flow DBD reactor can remove a significant amount of NO_X from a diesel engine exhaust.

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