

Discharge Plasma Treatment for NO_x Reduction from Diesel Engine Exhaust: A Laboratory Investigation

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ABSTRACT

A detailed study on the removal of oxides of nitrogen (NO_x) with and without the presence of carbonaceous soot in a stationary diesel engine exhaust was carried out using pulsed electrical discharges/catalyst/adsorbent processes. The processes were separately studied first and then the cascaded processes namely plasma-catalyst and plasma-adsorbent were examined. To investigate the effect of carbonaceous soot on the plasma treatment process, the filtered and unfiltered exhaust was treated by plasma separately. In the cascaded plasma-catalyst process, the plasma treating filtered exhaust was cascaded with a reduction catalyst V₂O₅/TiO₂ using ammonia as reducing agent and in the cascaded plasma-adsorbent process, the plasma treating unfiltered (raw) exhaust was cascaded with adsorbents (MS-13X/Activated alumina/Activated charcoal). The enhanced NO_x removal efficiency of plasma process in the presence of soot is identified, possible pathways are summarized and the results of the cascaded processes are discussed in detail.

Index Terms — Pulsed discharges, NO_x removal, discharge plasma, non-thermal plasma, engine emission, raw exhaust, plasma-catalyst, plasma-adsorbent, particulate treatment, Stationary emission source.

1 INTRODUCTION

REMOVAL of NO_x from diesel exhaust has been a challenge to researchers, as many conventional techniques such as catalysis, exhaust gas recirculation and other engine design modifications have failed to bring down the level of NO_x to mandatory limits put across various countries. In this context, electrical discharge plasma technique appears to be very promising [1]. Majority of the discharge-based studies were done using simulated pure gas mixtures and most of them were carried out at room temperatures. Though it is simpler to understand the detailed mechanisms and reaction pathways with simulated gas mixture treatment, it is very important to study the actual exhaust treatment using plasma reactor as the diesel exhaust consists of various components, which cannot simply be simulated using pure gases. Further, the diesel engine raw (unfiltered) exhaust contains more particulate matter as compared to petrol engine. The particulate matter present in the raw exhaust is comprised of dry carbon (soot) and volatile organic fraction (VOF), which is nothing but hydrocarbons (HC). Literature shows that the presence of hydrocarbons and carbonaceous soot in

diesel engine raw exhaust enhances the NO_x removal efficiency, when the raw exhaust is treated with discharge plasma [2–4].

It has been established that electric discharge plasma alone cannot remove NO_x from diesel exhaust [5, 6]. This demands discharge plasma to be combined with other aftertreatment techniques. Literature is abound with many works with hybrid plasma techniques [7–12].

In this paper, the NO_x removal studies were carried out for two cases. In the first case filtered diesel engine exhaust is treated with plasma and/or plasma assisted by a reduction catalyst. In the second case unfiltered diesel engine exhaust (raw exhaust) is treated with plasma and/or plasma-adsorbent technique. The effect of carbonaceous soot in enhancing the NO_x removal efficiency in the second case is discussed further. In the experiments involving catalyst or adsorbent, the exhaust was filtered before entering the catalyst or adsorbent.

The studies were carried out at different temperatures for the filtered exhaust case and for the unfiltered exhaust case the temperature was maintained at room temperature. The filtered exhaust treatment was conducted at no load and at 50% load, while the unfiltered exhaust treatment was conducted at no load condition.

2 EXPERIMENTAL SETUP

The experimental setup is shown in Figures 1a and 1b. The details of the setup are given below.

2.1 HIGH VOLTAGE PULSE SOURCE

This consists of a 0–25 kV high voltage ac system, a rectifier unit, a charging capacitor, a rotary spark gap (RSG) and the corona (plasma) as load. The hemispherical rotating electrode of the RSG was connected to a motor through an insulating rod. By changing the speed of the motor the frequency of the pulses applied to the plasma reactor can be controlled. Throughout the experiments, the frequency of the pulses was kept constant at 130 pulses per second (pps). The pulse voltage applied to the reactor was measured by means of a 150 MHz digital oscilloscope (DL1540, 200 MS/s, Yokogawa) connected through a 2000:1 voltage divider (EP-50K, 50 MHz, PEEC, Japan). The current was measured using a current probe (P6021, Tektronix) and the power was measured directly from the oscilloscope as the product of voltage and current waveforms.

2.2 DIESEL EXHAUST SOURCE

The diesel exhaust for the treatment was taken from a 4.4 kW diesel generator. The exhaust was treated under both no-load and 50% load conditions. An electric heater was connected as a load to the engine for carrying out the experiments at load condition. A part of the exhaust gas was made to pass through the treatment zone and the gas flow rate in the treatment zone was maintained at 4 L/minute.

2.3 PLASMA REACTOR (PR)

A dielectric barrier discharge reactor (referred to as plasma reactor) was employed in the present studies. The plasma reactor was a cylindrical glass tube (inner diameter 15 mm and outer diameter 17 mm) consisting of a stainless steel rod of thickness 1 mm as the inner electrode and aluminium foil wrapped over the glass tube as the outer electrode. The effective length of the reactor where discharge took place was 30 cm. The experiments involving plasma reactor were carried out at temperatures of 24, 100 and 150°C. For this, the reactor was placed in an electric furnace whose temperature could be controlled according to the requirements.

2.4 CATALYTIC REACTOR (CR)

The catalyst used was a commercially available V_2O_5/TiO_2 in the form of honeycomb (20 cells per square inch). The content of vanadium in the catalyst was 5 wt% and the apparent volume of the honeycomb was 31 cm^3 ($1.8 \times 1.8 \times 9.7 \text{ cm}^3$). This honeycomb structure was inserted in a glass tube of 26 mm inner diameter and 30 cm length, which formed the catalytic reactor. The catalytic

reactor was placed after the plasma reactor. Together, the combination is referred to as cascaded plasma-catalytic reactor. In this configuration, the plasma reactor was operated at room temperature whereas the catalytic reactor was operated at 100, 150 and 200°C.

2.5 ADSORBENT REACTOR (AR)

This is made of quartz glass tube of 15 mm diameter and effective length of 35 cm filled with the adsorbent beads such as activated charcoal/Molecular sieves MS-13X/activated alumina. This reactor was kept at room temperature.

2.6 FILTERING AND CONDITIONING SYSTEM (FCS)

This consists of two oil mist filters, one solid particulate filter and a moisture separator (UF10 SS, Ultra Filter India Ltd, India). The exhaust gas coming out of this unit has dew point of -23°C at atmospheric pressure. Proper care has been taken in the development of this conditioning system so as not to affect the sample gas components.

2.7 FT-IR UNIT

The measurement of NO_x, CO and other gaseous pollutants present in the diesel engine exhaust gas was carried out accurately using a multicomponent FTIR gas analyzer (DX-4010, Temet Instruments, Finland). The FTIR uses the CALCMET software to compute the concentrations of the components present in the sample gas from the absorbance spectrum.

3 RESULTS AND DISCUSSION

A 4.4 kW diesel generator was used for the experiments. Initially, before taking any measurement with FT-IR, zero calibration of FTIR was done by passing N₂ gas. Before treating the exhaust gas the concentrations of CO, total hydrocarbon (THC), NO_x, N₂O and aldehyde were measured. Table 1 shows the typical concentrations of the pollutants under no-load and 50% load conditions. In the table, NO_x means sum of concentrations of NO and NO₂. The concentrations of NO and NO₂ were measured individually and then added to get the NO_x concentration. The total hydrocarbons (THC) are the sum of various HCs present in the exhaust, which could be measured using

Table 1. Initial Concentrations of diesel engine exhaust components.

Components	No load	50% load
H ₂ O	0.3 vol%	0.3 vol%
CO ₂	2 vol%	4 vol%
O ₂	11 vol%	9 vol%
CO	560 ppm	720 ppm
NO _x (NO + NO ₂)	250 (210 + 40) ppm	710 (620 + 90) ppm
THC	95 ppm	200 ppm
Aldehydes	40 ppm	75 ppm

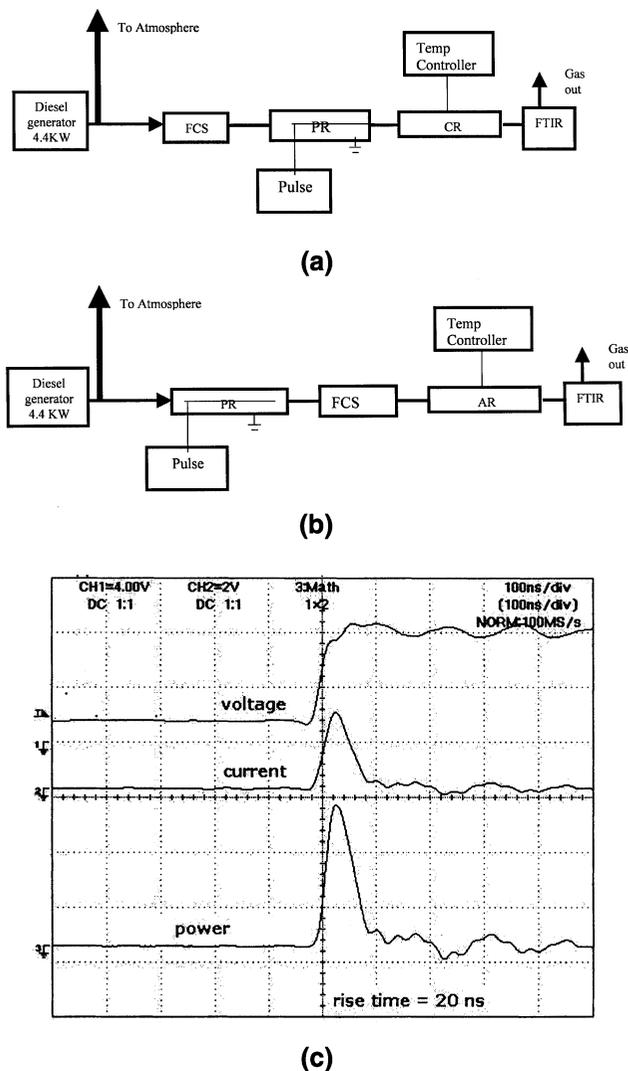


Figure 1. a, Experimental setup for diesel engine filtered exhaust treatment; b, Experimental setup for diesel engine unfiltered exhaust treatment; c, Waveforms of voltage, current and power. FCS, Filtering and conditioning system; PR, Plasma reactor; CR, Catalytic reactor AR, Adsorbent reactor.

FTIR analyzer. THC in the present study includes methane (CH₄), ethylene (C₂H₄), acetylene (C₂H₂), butane (C₄H₁₀), propene (C₃H₆), benzene (C₆H₆), and toluene (C₇H₈). Aldehydes included formaldehyde and acetaldehyde.

In the present paper, all the results were presented in terms of specific energy density. At first, the discharge energy per pulse was measured as the integration of the product of voltage and current waveforms as shown in Figure 1c. In the figure Trace1, Trace2 and Trace3 respectively refer to voltage, current and power. Multiplying the discharge energy per pulse by the pulse repetition rate gave the average discharge power. The energy density was calculated as the ratio of average discharge power to the gas flow rate. The results were first presented for the filtered exhaust case covering plasma technique and cas-

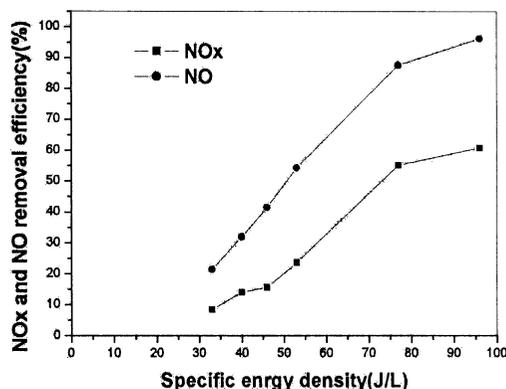


Figure 2. Variation of NO_x and NO removal efficiencies with respect to specific energy density, in case of plasma reactor treating filtered exhaust at room temperature under no load condition.

caded plasma-catalytic technique, then the results were shown for the unfiltered (raw) exhaust case, discussing plasma, adsorbents and cascaded plasma-adsorbent techniques.

3.1 FILTERED EXHAUST TREATMENT

3.1.1 PLASMA REACTOR (PR)

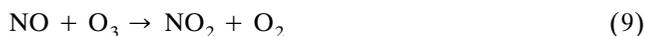
3.1.1.1 NO_x/NO REMOVAL AT ROOM TEMPERATURE UNDER NO LOAD CONDITION

Figure 2 shows the variation of NO_x and NO removal efficiencies with respect to output specific energy density. Both NO_x and NO removal efficiencies increase with increase in energy density. For instance, at an energy density of 66 J/L, NO_x and NO removal efficiencies of 45 and 75%, respectively are recorded. Possible reaction pathways responsible for NO_x and NO removal can be summarized as below [2, 3, 6, 13, 14].

NO_x removal reactions

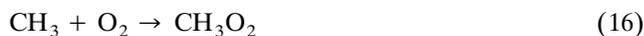


NO-NO₂ conversion reactions involving O/O₃ radicals



NO-NO₂ conversion reactions involving HC and O radicals





NO-NO₂ conversion reactions involving HC and OH radical



Diesel exhaust being an oxygen rich exhaust, contains large number of O radicals which enable efficient oxidation of NO to NO₂, thus making the plasma environment NO₂ rich in case of filtered exhaust plasma treatment [1, 15]. NOx removal in this environment is favored mainly by methoxy radical induced reactions (3), (4) and (5) [14].

3.1.1.2 EFFECT OF HIGHER EXHAUST TEMPERATURE AND ENGINE LOADING ON NO_x/NO REMOVAL

Figure 3 shows effect of exhaust temperature on NO removal efficiency under no load condition. With increase in output energy density, the NO removal increases at all exhaust temperatures and at a given energy density, the NO removal decreases with increase in exhaust temperature. At an energy density of 86 J/l, the NO removal efficiency at 24, 100 and 150°C was 90, 69 and 44%, respectively. This indicates that, at high temperatures, more energy input is required to bring about high NO removal efficiencies. The decrease in NO removal efficiency with temperature can be attributed to the following:

(a) The rate coefficient of oxidation reaction (7) decreases with increase in temperature thereby reducing the efficiency of NO conversion to NO₂.

(b) Depletion of ozone at increased temperature which otherwise plays an important role in NO oxidation (reaction 9).

(c) At increased temperature, the NO₂ gets reduced back to NO (NO₂ + O → NO + O₂).

(d) Many hydrocarbons, which enhance the oxidation of NO at room temperature, may not do the same at increased temperatures. This is because, the rate coefficients of reactions involving the release of active hydrocarbon species decreases at increased temperatures [16].

The NO removal efficiencies at different temperatures for 50% load exhaust are shown in Figure 4. The room

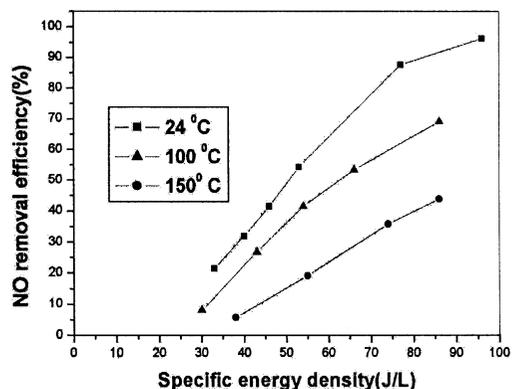


Figure 3. NO removal efficiencies at different exhaust temperatures in case of plasma reactor treating filtered exhaust under no load condition.

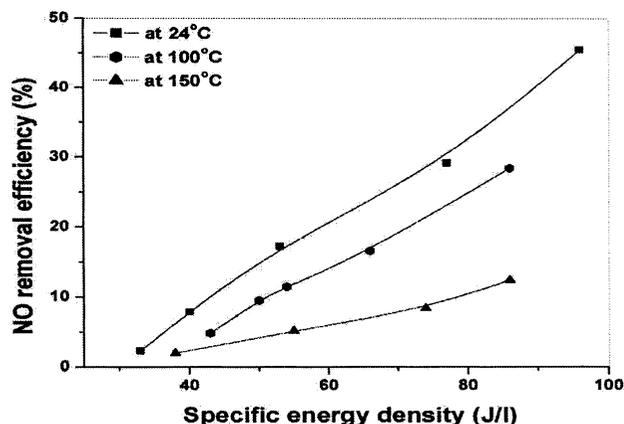


Figure 4. NO removal efficiencies at different exhaust temperatures in case of plasma reactor treating filtered exhaust under 50% load condition.

temperature treatment gave rise to maximum NO removal efficiency of 45%. This value was less than half of the removal efficiency obtained when the exhaust was treated under no load conditions. This was because of high NO initial concentration under load condition (620 ppm). NO removal efficiency decreases with increase in initial concentration of NO [17]. For a given electric field, the number of radicals produced is constant. However, the ratio of active radicals with respect to NO molecules decreases with increase in initial NO concentration thereby decreasing the NO removal. Similar to the results obtained under no-load conditions, the NO removal efficiencies decreased at increased temperatures under load conditions. At temperatures, 100 and 150°C the NO removal efficiencies obtained were only 28 and 12%, respectively.

Figure 5 shows the NOx removal efficiencies obtained at various temperatures when the exhaust was treated under no load conditions. At room temperature, at an energy density of 96 J/L, the NOx removal efficiency was 60%, while at 100 and 150°C, it was 33 and 21%, respectively. The reason for this decrease was as explained in

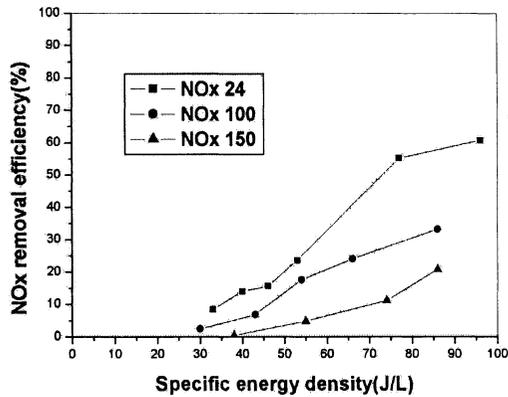


Figure 5. NO_x removal efficiencies at different exhaust temperatures in case of plasma reactor treating filtered exhaust under no load condition.

the previous section. When the exhaust under 50% loads was treated, the NO_x removal decreased further. At room temperature, the NO_x concentration reduced to 535 ppm from an initial concentration of 710 ppm. This amounted to a removal efficiency of 25%. At 100 and 150°C, the NO_x removal efficiencies were 20 and 10%, respectively.

3.1.2 CASCADED PLASMA-CATALYTIC REACTOR (CPCR)

The catalytic reactor, with V₂O₅/TiO₂ as the catalyst, was placed after the plasma reactor. The catalyst V₂O₅/TiO₂ is a SCR catalyst, which uses ammonia as its reducing agent. Ammonia was added in a ratio of 1:1 to NO_x. In this system, the plasma reactor was operated always at room temperature whereas the catalytic reactor was operated at temperatures of 100, 150 and 200°C. The reason for operating plasma reactor at room temperature was that the NO removal efficiency obtained was very less at increased temperatures especially under no load conditions. As can be seen in the coming section, the NO removal (NO₂ production) using plasma reactor plays a very

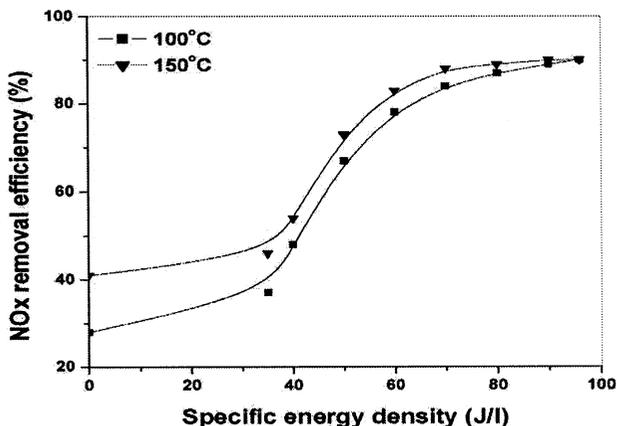


Figure 6. NO_x removal efficiencies at different exhaust temperatures in case of cascaded plasma-catalytic reactor under no load condition.

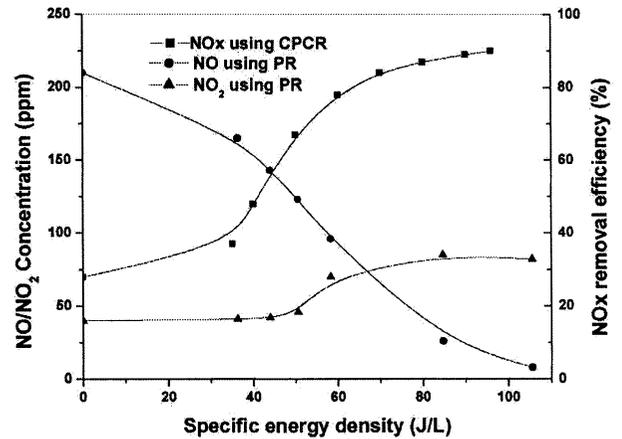


Figure 7. NO and NO₂ concentrations obtained using PR along with the NO_x removal efficiency obtained using CPCR under no load condition. PR at room temperature and CPCR at 100°C.

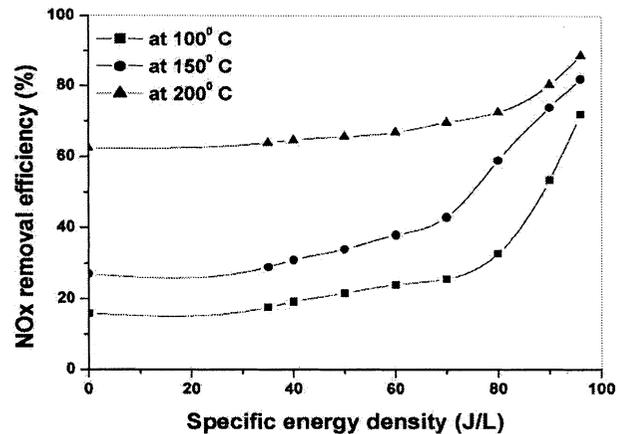


Figure 8. NO_x removal efficiency at different exhaust temperatures in case of cascaded plasma-catalytic reactor under 50% load condition.

important role in helping the catalysis process to take place effectively even at temperatures below 200°C. If hydrocarbons were added to the exhaust stream, the plasma reactor could be operated at temperatures above 24°C even under load conditions. However, in the present paper work, the hydrocarbons were not added.

In all the figures pertaining to CPCR (Figures 6-9), the zero energy input (0 J/L) corresponds to the catalyst performance alone without plasma being operated. Individual figure is now being discussed. Figure 6 shows the NO_x removal efficiencies obtained under no-load conditions of the exhaust. At 100°C, the CPCR achieved a maximum NO_x removal efficiency of 90%. The performance of catalyst alone at 100°C in removing NO_x was 27%. As the energy input to plasma reactor increases, the NO_x removal increases and reaches 84% at an energy density of 70 J/L. Beyond this energy input, the increase in NO_x removal becomes slower and finally gets stabilized to 90% at 96 J/L. At 150°C, similar trends were observed. The

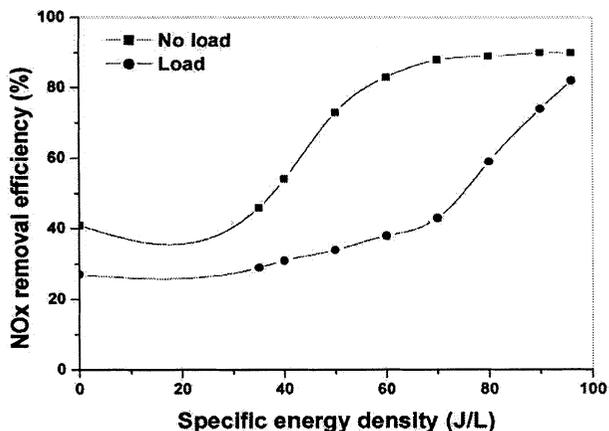
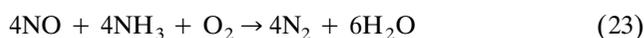


Figure 9. Comparison of NOx removal efficiencies in case of cascaded plasma-catalytic reactor under 50% load and no load conditions at an exhaust temperature of 150°C.

maximum NOx removal achieved was same as that at 100°C. The NOx removal efficiency at 200°C is not shown because the NOx removal by catalyst alone was 80% and that there was no necessity of combining it with plasma. The NOx removal using this CPCR system can be explained as below.

According to [18], the SCR catalysis is more pronounced when the NOx in the exhaust stream consists of equal amounts of NO and NO₂. Since the plasma reactor oxidizes NO to NO₂, the exhaust entering the catalytic reactor consists of both NO and NO₂. With the increase in NO₂ concentration entering the catalytic reactor, the NOx removal increases. Ammonia in presence of the catalyst reduces NO and NO₂ mainly through the following reaction,



Reaction (23) is known to take place at high temperatures such as above 200°C whereas reaction (24) can take place below 200°C also. The overall reaction can be represented as

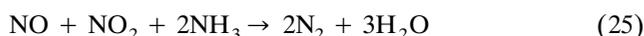


Figure 7 presents NO and NO₂ concentrations obtained using PR along with the NOx removal efficiency obtained using CPCR, under no load condition. The concentrations of NO and NO₂ at the outlet of the PR become equal for an energy input of around 60 J/L. It is observed that, the NOx removal efficiency obtained using CPCR at an energy density of 60 J/L was very high suggesting that equal NO and NO₂ concentrations help in obtaining high NOx removal.

When the exhaust was treated under 50% load conditions using CPCR, the NOx removal efficiency was nearly

unaffected up to an energy density of 80 J/L at all temperatures, as shown in Figure 8. This was because the NO to NO₂ conversion using plasma reactor was less up to an energy density of 80 J/L. Under 50% load condition, the NO removal efficiency obtained using the plasma reactor alone was only 30% at 80 J/L and 45% at 96 J/L (see Figure 4) at room temperature. The corresponding NOx removal efficiency obtained in case of CPCR 100°C, are 30% and 72%, respectively. Similar trends were observed at temperatures 150 and 200°C. The NOx removal efficiencies obtained at 150 and 200°C using CPCR were 82 and 88%, respectively.

The NOx removal efficiencies obtained under 50% load conditions were lesser than those under no-load conditions because of high initial NO concentrations. Figure 9 compares the NOx removal efficiencies under load and no-load conditions at an exhaust temperature of 150°C. Under no-load conditions, it was found that at less energy input, NOx removals achieved were high compared to those under load conditions. For example, at an energy input of 70 J/L, the NOx removal under load conditions was only 43% as against 88% under no-load conditions. At maximum energy input of 96 J/L, the respective NOx removal efficiencies under no-load and 50% load conditions achieved became 90 and 82%, respectively.

3.2 UNFILTERED (RAW) EXHAUST TREATMENT

In this case, all experiments were carried out at room temperature under no load condition.

3.2.1 PLASMA REACTOR (PR)

Figure 10 gives the variation of NOx removal efficiency with respect to energy density. The NOx removal efficiency increases with increasing energy density [1, 19]. Further, comparing with Figure 2, at a given energy density, NOx removal efficiency was higher with plasma treating raw exhaust than that with treating filtered exhaust

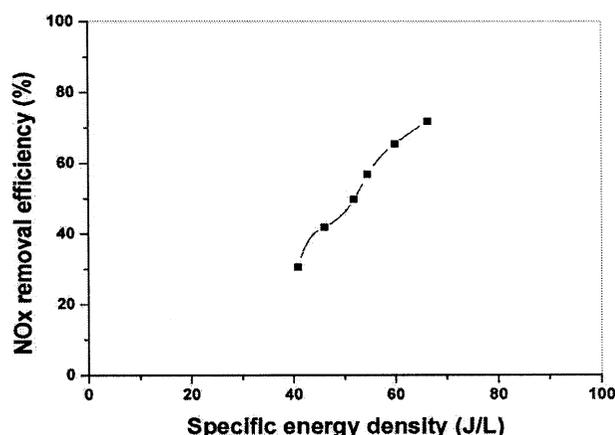


Figure 10. NOx removal efficiency in case of plasma treating unfiltered (raw) exhaust at room temperature under no load condition.

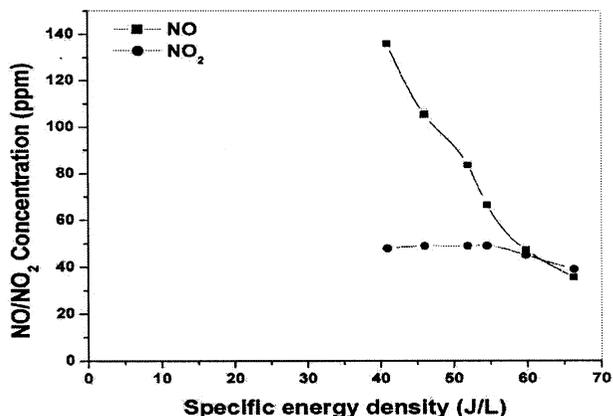
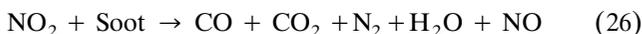


Figure 11. NO and NO₂ concentrations obtained using plasma reactor treating raw exhaust at room temperature under no load condition.

(ex: at 66 J/L, raw exhaust plasma treatment gives NO_x removal efficiency of 71%, while it was only 45 % with filtered exhaust plasma treatment). Further, in contrast to Figure 7, in raw exhaust treatment the NO₂ concentration remains almost constant and NO concentration decreases with increasing energy density as shown in Figure 11. This can be due to the following reaction pathways:

In the presence of soot, the key radicals (O/OH) responsible for NO – NO₂ conversion and hydrocarbon oxidation are getting depleted by reacting with carbonaceous soot and as a result, the reactions (10) to (22) (section 3.1.1) are less probable. Hence, NO₂ formation is affected. The partly converted NO₂ reacts with soot by the following reaction [2]:



As a result, NO₂ concentration will remain almost constant. Further, NO may be reduced to N₂ as the reactions 1 and 2 (section 3.1.1) appear to be more probabilistic in the depleted O and OH environment. Therefore, total NO_x is reduced to a value much less than that observed in filtered exhaust case [3, 20, 21, 22].

3.2.2 ADSORBENT REACTOR (AR)

Studies were conducted using three commercial adsorbents namely, activated charcoal, MS-13X and activated alumina. The NO_x removal efficiency with all the three adsorbents is shown in Figure 12. All the adsorbents show a marginal saturation effect after 30 minutes of use. The NO₂ molecules having a larger molecular weight compared to NO molecules show a higher degree of adsorption [12]. The NO molecules show a lower degree of adsorption in case of all the three adsorbents. Further, activated charcoal exhibits superior adsorption property compared to other two adsorbents because of its large internal surface area.

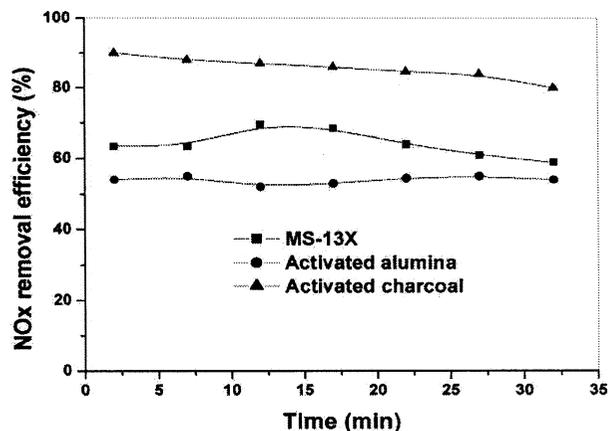


Figure 12. NO_x removal efficiency using adsorbents at room temperature under no load condition.

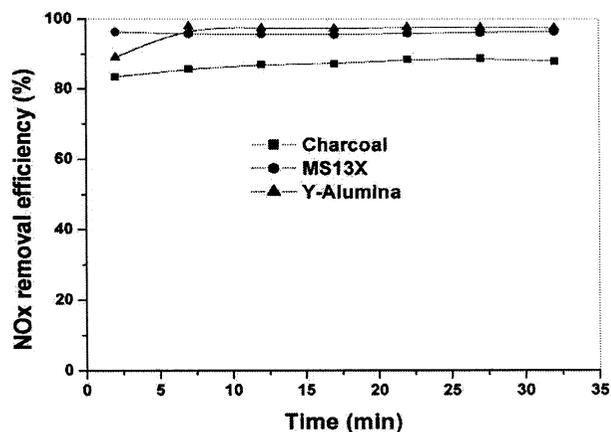


Figure 13. NO_x removal efficiency in case of plasma-associated adsorption at room temperature under no load condition.

3.2.3 CASCADED PLASMA-ADSORBENT REACTOR (CPAR)

The adsorbent reactor packed with adsorbent beads was placed after the plasma reactor. It should be noted that in all the experiments the plasma reactor was operated at a constant discharge energy density of 60 J/L corresponding to a voltage of 22 kV.

Figure 13 shows time dependent NO_x removal efficiency for the three adsorbents associated by plasma. For a given adsorbent, the plasma-associated adsorption exhibits superior NO_x removal characteristics compared to the individual processes- plasma/adsorption [8, 14]. At any given time, cascaded plasma-adsorbent reactor (any adsorbent), exhibits very high NO_x removal (96%). Owing to high NO_x removal characteristic (71%) of plasma treating raw exhaust, the exhaust entering an adsorbent contains lesser concentration of NO and NO₂ molecules, as a result the type of adsorbent plays a little effect on NO_x removal. Hence, cascaded plasma-adsorbent reactor configuration gives almost same NO_x removal efficiency irrespective of type of adsorbent used.

4 CONCLUSIONS

STUDIES were conducted on filtered and unfiltered (raw) stationary diesel engine exhaust using different aftertreatment techniques involving a combination of plasma, catalyst and adsorbents. The major inferences drawn from this work are summarized as below:

Presence of carbonaceous soot in the diesel engine raw exhaust enhances NO_x removal capability of a plasma process. It directly reacts with NO₂ and decreases the NO_x concentration. In the filtered exhaust treatment, cascaded plasma-catalytic reactor performed better than catalytic reactor (V₂O₅) alone for temperatures below 200 °C. In case of cascaded plasma-catalytic reactor, at an energy density of 60 J/L, at which concentrations of NO and NO₂ become equal, a very high NO_x removal efficiency was observed. The activated charcoal exhibits a superior NO_x adsorption property compared to MS-13X and activated alumina. For a given adsorbent, the plasma-associated adsorption exhibits superior NO_x removal characteristics compared to the individual processes. The plasma-associated adsorption technique, exhibiting a very high NO_x removal, can be a viable alternative aftertreatment technique, to plasma assisted selective catalytic technique. The authors are currently developing a prototype of hybrid plasma-adsorbent system for NO removal in engine exhausts.

ACKNOWLEDGMENTS

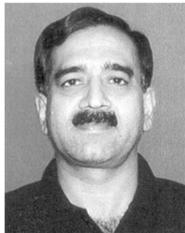
The authors wish to thank M/s Zeolites, India, for supply of adsorbent samples. This research was partially financed by University Grants Commission, India, under the Science and Technology Infrastructure Development Programme (COSIST/ASIST, 2001–05).

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