

A Laboratory Investigation of Pulsed Discharged Based Techniques for Engine Exhaust Treatment

- Effect of Exhaust Nature and Operating Conditions

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Abstract—A detailed investigation on the removal of pollutants (NO_x, aldehydes and CO) from the exhaust of a stationary diesel engine is carried out using pulse discharge plasma associated with adsorbent-catalyst techniques (PAC). The objective of the study is to explore the effect of the exhaust nature, i.e. filtered or raw, and operating conditions on the pollutant (NO_x, CO and aldehydes) removal process. In this study the exhaust treatment was carried out in two stages. In the first stage, the exhaust was treated with single step PAC and in the second stage with double step PAC. To study the effect of exhaust nature, in each stage the experiments were carried out with filtered and unfiltered (raw) exhaust. Further, to study the effect of operating conditions, in each stage, the experiments were carried out at different temperatures (up to 400°C), different engine loading and flow rate. The effectiveness of the technique with regard to NO_x, CO removal and by-product reduction was discussed. Finally, a comprehensive comparison of the single step PAC and double step PAC techniques has been made and results were discussed.

Keywords—Pulse discharge plasma, diesel engine exhaust, single step PAC, double step PAC, pollutant removal, by-products formation, plasma based techniques

I. INTRODUCTION

Controlling emissions from combustion engines particularly from diesel driven ones is a challenge to the researchers across the globe. In the case of diesel engines despite the modifications in engine design and improvement in after treatment technologies, large amount of NO_x and CO continue to emit and attempts to develop new catalysts to reduce these pollutants have been so far less successful.

The electrical discharge plasma (non-thermal plasma) is a prominent non-conventional technique, which can produce chemically active species that can facilitate the removal of NO_x and other pollutants within diesel exhaust [1-10].

Further, plasma promotes catalysis and adsorption when it is combined with a catalyst and an adsorbent. Plasma associated catalysis and adsorption are gaining lot of importance [11-23]. However, majority of the research work on actual diesel engine exhaust has been done at exhaust temperatures higher than 150°C making use of proprietary catalysts with the use of additional hydrocarbons. The results reported have limitations with regard to pollutant removal efficiency, byproduct formation, pollutant initial concentration, energy consumption and operating temperature window.

In the present work, a detailed study on the removal of pollutants (NO_x, aldehydes and CO) from the exhaust of a Stationary diesel engine was carried out using barrier discharge hybrid plasma techniques. The objective of the study is to explore the effect of the exhaust nature, i.e. filtered or raw, and operating conditions on the pollutant

(NO_x, CO and aldehydes) removal process. In this study the exhaust treatment was carried out in two stages. In the first stage, the exhaust was treated with single step Plasma based techniques and in the second stage with double step Plasma based techniques. To study the effect of exhaust nature, in each stage the experiments were carried out with filtered and unfiltered (raw) exhaust. Further, to study the effect of operating conditions, in each stage, the experiments were carried out at different temperatures (up to 400°C), different engine loading and flow rate. The effectiveness of the technique with regard to NO_x, CO removal and by-product reduction was discussed. Finally, a comprehensive comparison of the single step and double step plasma based techniques has been made and results were discussed.

II. EXPERIMENTAL SETUP

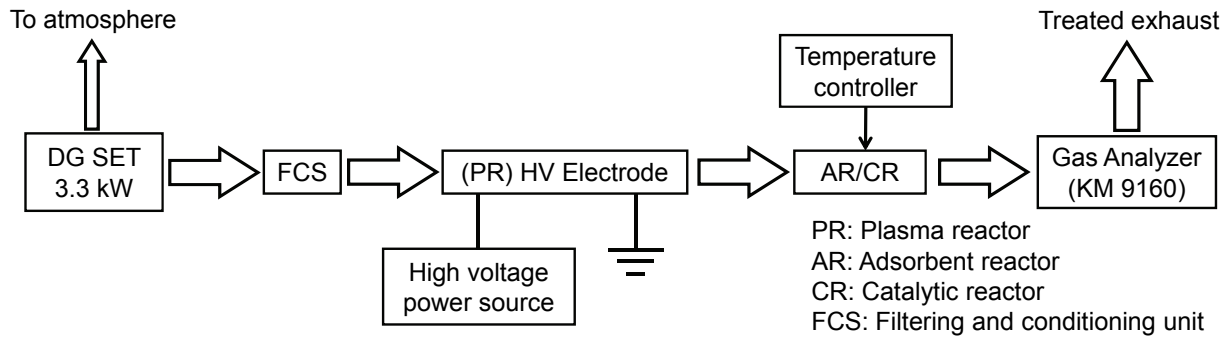
The schematic of the diesel engine exhaust treatment setup is shown in Fig. 1. Fig. 1(a) represents the schematic for single step plasma based technique while Fig. 1(b) represents the schematic for double step plasma based techniques.

A 30 kV pulse source was used in the studies. Throughout the experiments, the frequency of the pulses was kept constant at 100 pps (pulses per second). The pulse voltage applied to the plasma reactor was measured by means of a 150 MHz digital oscilloscope (DL1540, 200MS/s, Yokogawa) connected through a 2000:1 voltage divider (EP-50K, 50MHz, PEEC, Japan). The current was measured using a current probe (P6021, Tektronix).

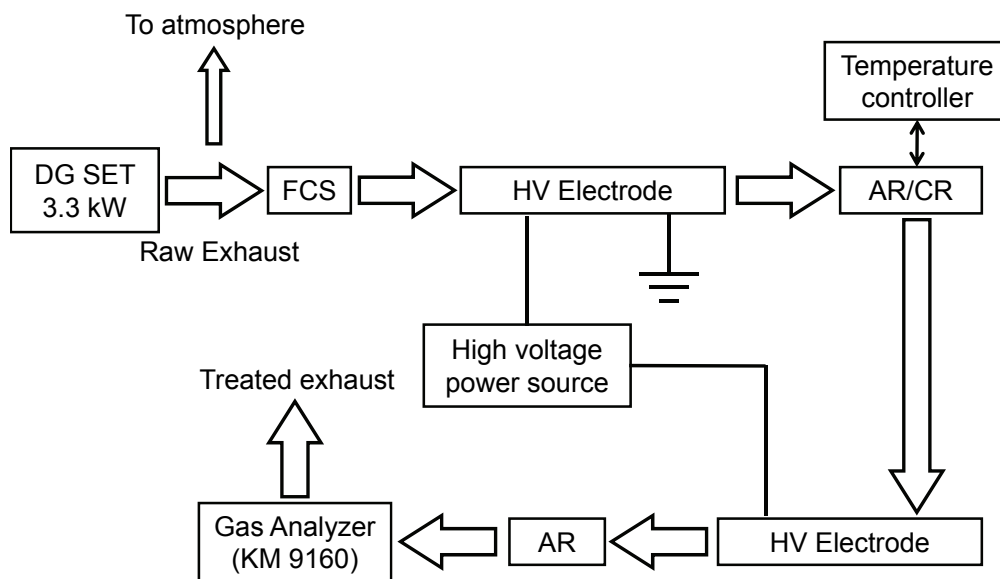
A 3.3 kW diesel engine was used as the exhaust source. The whole of the exhaust from the engine was not treated in view of infrastructure limitation in the laboratory. Further, as our objective is to examine the

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(a) Single step plasma/plasma hybrid technique



(b) Double step plasma/plasma hybrid technique

Fig. 1. Diesel engine exhausts treatment using plasma/plasma hybrid technique.

underlying principle involved in the exhaust treatment, only a part of the main exhaust from the engine was treated and the exhaust flow rate was controlled and varied from 4 lpm to 8 lpm.

A dielectric barrier electric discharge reactor (referred to as plasma reactor (PR)) 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 aluminum 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 room temperature.

Two types of non-conventional commercially available Catalysts were used. The catalysts used were red mud and activated alumina catalyst. Both the catalysts were in the form of pellets. The catalysts were placed inside the quartz glass tube of 30 cm length and 15 mm diameter. This is referred to as catalytic reactor (CR).

Two types of non-conventional commercially available adsorbents were used. The adsorbents used

were molecular sieves MS 13X and activated alumina. The adsorbents in the form of beads were placed inside quartz glass tube of 15mm diameter and effective length of 30 cm. This is called adsorbent reactor (AR). The adsorbent reactor was operated at room temperature.

In the single step plasma catalyst hybrid configuration the catalytic reactor was placed after the plasma reactor and in the double step plasma catalyst hybrid configuration another plasma reactor is included after the single step configuration. In both these configurations, the plasma reactors were operated at room temperature, whereas the catalytic reactor was operated at temperatures varying from room temperature to 400°C.

In the single step plasma adsorbent hybrid configuration the adsorbent reactor was placed after the plasma reactor and in the double step plasma adsorbent hybrid configuration another single step plasma adsorbent hybrid configuration is included in series with the first single step configuration. In these configurations, both the plasma reactors and adsorbent reactors were operated at room temperature.

In the experiments with filtered exhaust, filtering of the exhaust was done first, using filtering & conditioning unit (FCS). The filtered exhaust was then allowed to enter the treatment zone. The exhaust gas was made to pass through a tube containing steel wool, in order to filter out oil mist and macro-sized particulate matter. The exhaust was then passed through filtering and conditioning system (FCS). The FCS consists of three filters and a moisture separator. The function of the FCS is to filter out the carbonaceous soot, any coarse particles, oil mists and water from the exhaust gas. Proper care has been taken in the development of this conditioning system so as not to affect the sample gas components. In the experiments with Raw exhaust, the exhaust from the engine was taken directly to the plasma reactor and then the filtered exhaust was allowed to pass through the catalytic// Adsorbent reactor

The measurement of NO_x, and other gaseous pollutants present in the diesel engine exhaust gas was carried out accurately using a QUINTOX KM 9160, Kane International UK gas analyzer.

III. RESULTS AND DISCUSSION

Before treating the exhaust gas, the concentrations of CO, CO₂, NO, NO₂, NO_x, O₂ and aldehydes were measured. Table I shows the typical concentrations of the pollutants under 0% load and 27.27% load conditions.

In Table I, 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. Aldehydes included formaldehyde and acetaldehyde.

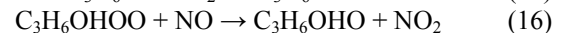
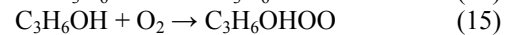
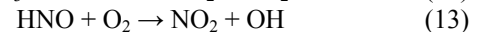
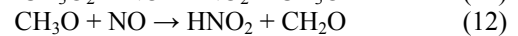
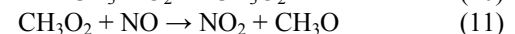
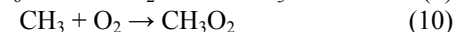
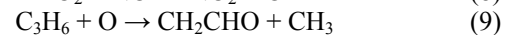
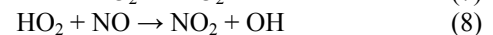
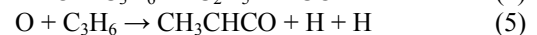
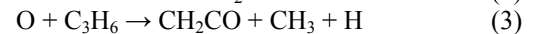
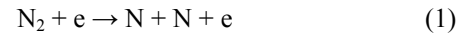
In the present paper, the results were presented in terms of specific energy density and pulse voltage in kV. The energy density was calculated as the ratio of average discharge power to the gas flow rate. The results were first presented for the plasma process, then for plasma catalyst hybrid process and finally for plasma adsorbent hybrid process.

TABLE I
INITIAL CONCENTRATION OF POLLUTANTS/COMPONENTS
PRESENT IN DIESEL ENGINE EXHAUST

Main pollutants	0% Load (No load)	27.27% Load
CO ₂	0.10%	0.60%
CO	323ppm	229ppm
NO	110ppm	257ppm
NO ₂	30ppm	69ppm
NO _x	140ppm	326ppm
Aldehydes	50ppm	80ppm
O ₂	20.70%	20.50%

A. Plasma Process

Both filtered and unfiltered diesel exhaust was treated by the plasma process. The basic NO_x conversion reactions involving N, HC, O, OH radicals which take place in the plasma are given below:



In the case of filtered diesel exhaust treatment, as we are removing water using the filter and conditioning unit there will not be significant effect of OH radicals on NO/NO₂ conversion. In the case of unfiltered diesel exhaust treatment (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, NO₂ concentration will remain almost constant. Further, NO may be reduced to N₂ as the reactions 1 and 2 appear to be more probabilistic in the depleted O and OH environment [24].

In this section, both single step and double step plasma processes were discussed. Fig. 2 shows the variation of NO_x removal efficiency for different pulse voltages at various exhaust flow rates under no load in the case of single step plasma process. The flow rate

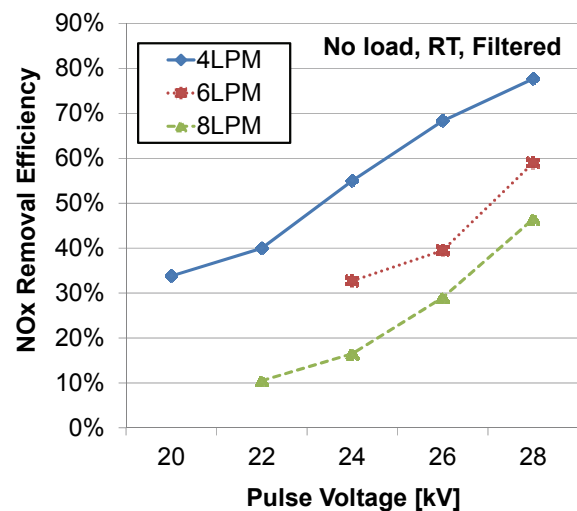


Fig. 2. Effect of flow rate on single step plasma.

considerably affects the NO_x removal which has to be taken care of when treating large flow rates. For this purpose the exhaust was treated by two step plasma and the effect of flow rate on NO_x removal is shown in Fig. 3. It is seen that for a given pulse voltage the NO_x removal is not much affected by the flow rate. This could be attributed to the increased exhaust resident time in the case of double step plasma process. This result is important while configuring the plasma reactors to handle large flow rates in practical situations.

Due to limitation in the present laboratory set up we are able to carry out the experiments at lower flow rates. However we aim to carry out the experiments at higher flow rates and higher SV in future.

Fig. 4 shows the superior NO_x removal performance of the double step plasma processes at energy density of 60 J/L under no load at a flow rate of 4 lpm. This confirms viability of a multistep plasma processing technique particularly at higher exhaust flow rate.

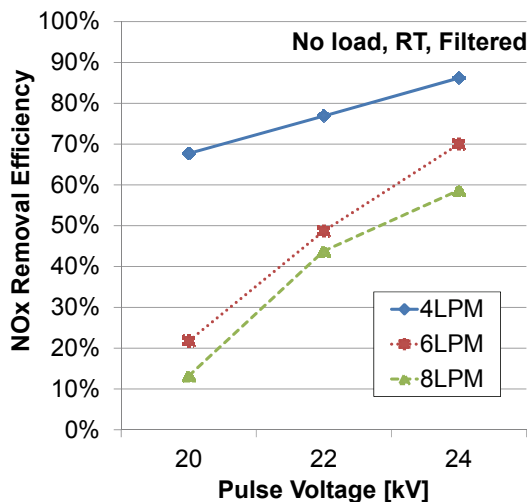


Fig. 3. Effect of flow rate on double step plasma.

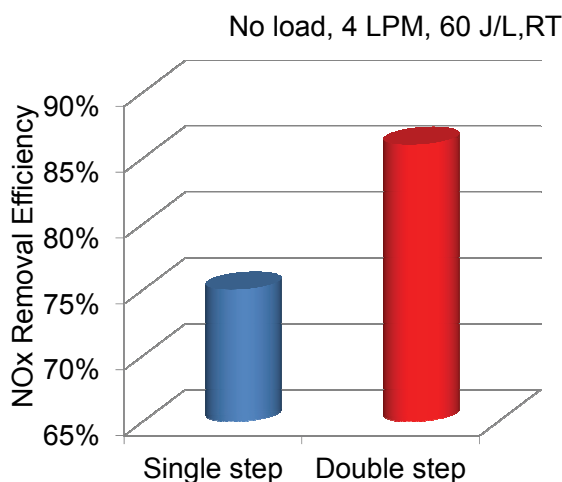


Fig. 4. Comparison of single step and double step plasma.

B. Catalyst Process

In this section two non-conventional catalysts Red mud and Activated Alumina in the form of pellets were used. It has been established that the NO_x removal increases with temperature for red mud [25], while it decreases with temperature for activated alumina [26]. This is an interesting behavior where the Aldehydes present in the diesel exhaust are just sufficient to activate red mud but not activated alumina. The excess Aldehyde requirement for activated alumina activation will be discussed in the next section on plasma assisted catalyst hybrid technique.

Red mud is generated as a waste during the processing of bauxite, the most common ore of aluminum. It is a by-product of bauxite processing through Bayer process. Red mud mainly contains a mixture of oxides of Fe, Al, Ti and smaller amounts of Si, Ca and Na. The main constituents of red mud include Fe₂O₃, Al₂O₃, SiO₂, TiO₂, Na₂O, CaO, MgO and a number of minor constituents like K, Cr, V, Ni, Cu, Mn, Zn etc. Generally ferric oxide (Fe₂O₃) is the major constituent of red mud and gives it its characteristic brick red colour. The surface area of red mud powder lies between 20-30 m²/g. Red mud has a fine particle size distribution with 90% by volume below size of 75 micron, and high surface area.

Fig. 5 shows the effect of load on the NO_x and CO removal by red mud catalyst for different temperatures. Various reaction pathways have been discussed for NO_x and CO removal [24]. However the exhaust concentration is substantially high at load condition which reduces the activity of the catalyst in turn resulting in reduced NO_x and CO removal under load condition.

Fig. 6 gives the effect of flow rate on NO_x and CO removal in red mud catalyst. It is seen that at high flow rates, both NO_x and CO removal decreases and further the effect of flow rate is more pronounced with regard to CO removal than NO_x removal. This can be due to reduced activity of the reactions involving CO [25].

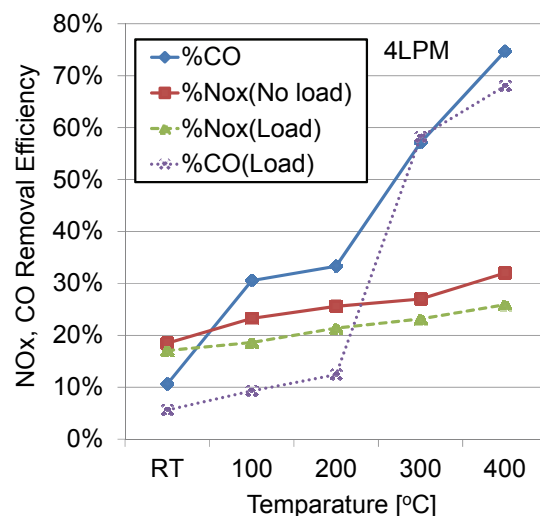


Fig. 5. Effect of load on NO_x and CO in red mud catalyst removal.

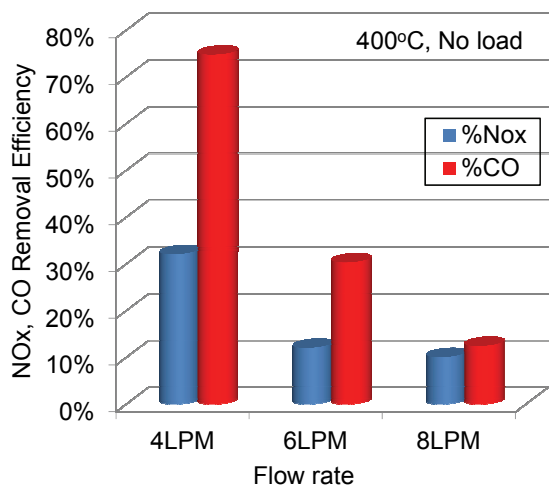


Fig. 6. Effect of flow rate on NOx and CO removal in red mud catalyst

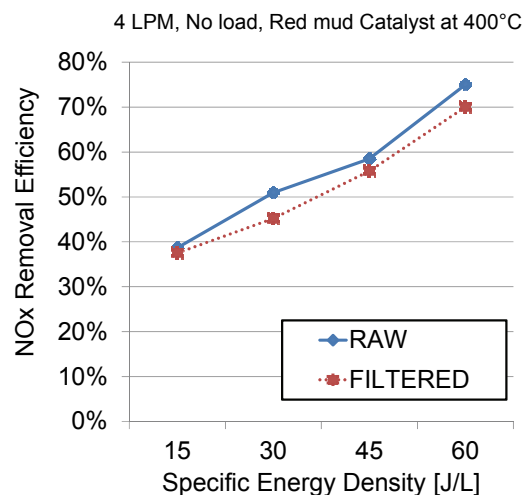


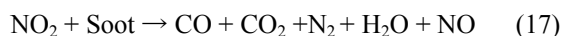
Fig.7. Effect of exhaust nature on single step plasma assisted catalytic process.

C. Plasma Assisted Catalyst Hybrid Process

In this section plasma assisted catalyst hybrid process was investigated in single step and double step configuration using red mud and activated alumina catalysts. Both filtered and unfiltered diesel exhaust were treated. In the filtered exhaust treatment the exhaust is filtered before entering the plasma and catalyst reactors. However in the unfiltered exhaust treatment the raw exhaust is allowed to pass through the plasma reactor and is then filtered before entering the catalyst reactor. Hence in both the cases, filtered exhaust only enters the catalyst reactor and there is no possibility of soot deposition on the surface of the catalyst.

The NOx removal performance of plasma process alone is different in the filtered and unfiltered exhaust treatment.

(i) In the presence of soot (unfiltered exhaust), 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, NO₂ formation is affected. The partly converted NO₂ reacts with soot by the following reaction



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

(ii) In the filtered exhaust case the conversion from NO to NO₂ is significantly high in the plasma reactor where at energy density 60 J/L the NO₂ was increased to 110ppm from an initial concentration of 30ppm. However the total NOx reduction was low due to oxidizing environment of plasma. Further the O radicals

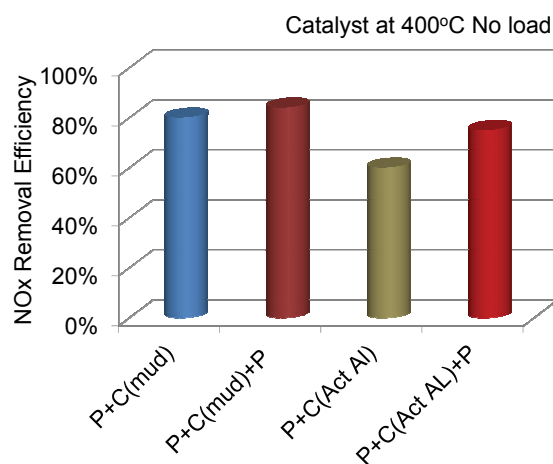


Fig. 8. Comparison between single step and double step plasma assisted catalytic process.

produced in the plasma react with hydrocarbons forming aldehydes.

However, when plasma is associated with red mud catalyst the overall NOx removal performance under filtered and unfiltered conditions remain almost the same as shown in Fig. 7. This can be explained as below.

In the case of filtered exhaust treatment, NO₂ which was increased to 110ppm in the plasma is getting reduced to 17ppm at downstream end (at the exit of plasma-catalyst reactor), which is less than initial concentration. This is because the aldehyde produced in the plasma act as reducing agent for NO₂ reduction in the catalyst. Thus the decreased NOx removal by plasma is compensated by increased NOx removal by catalyst.

In the case of unfiltered exhaust treatment due to the presence of soot there is no significant increase in NO₂ concentration in the plasma and NOx removal is better. However as the aldehyde production is less, the NO₂ reduction is not effective in the catalyst. Hence increased NOx removal by plasma is compensated by decreased NOx removal by catalyst.

Fig. 8 gives comparison between single step and double step plasma assisted catalyst process with regard to NO_x removal. In single step the hybrid process with red mud as catalyst performs better compared to activated alumina as catalyst which can be attributed to better catalytic action of red mud. However in the case of double step hybrid process the type of catalyst has a little effect on the NO_x removal which can be due to a major contribution from the plasma process in the double step hybrid configuration.

D. Plasma Assisted Adsorbent Hybrid Process

In this section plasma assisted adsorbent hybrid process was investigated in single step and double step configuration using MS13x and activated alumina adsorbents.

The superior performance of plasma assisted adsorbent hybrid process has been well established [15]. Fig. 9 gives comparison between single step and double step plasma associated adsorbent process with regard to NO_x removal. In single step the hybrid process with MS13X as adsorbent performs better compared to activated alumina as adsorbent which can be attributed to

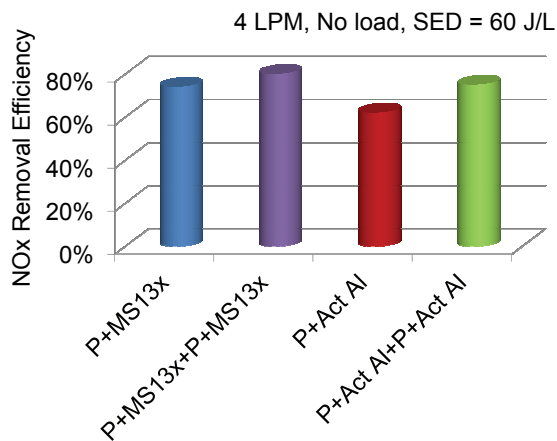


Fig. 9. Comparison of single step and double step plasma assisted adsorbent process.

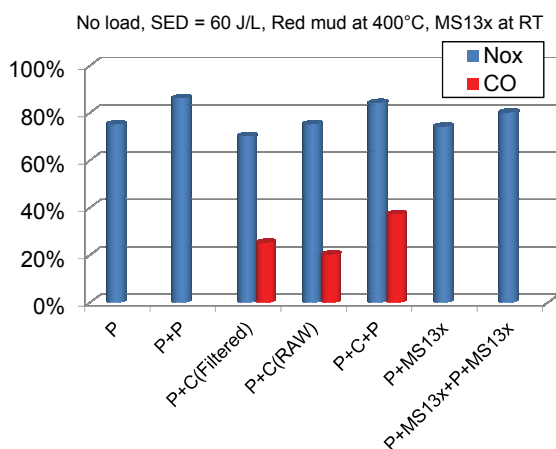


Fig. 10. Performance of different types of techniques for removal of NO_x and CO.

better adsorbent action of MS13X [26, 15]. However in the case of double step hybrid process the type of adsorbent has a little effect on the NO_x removal which can be due to a major contribution from the plasma process in the double step hybrid process.

Fig. 10 shows performance of different types of techniques used for NO_x/CO removal from diesel exhaust.

IV. CONCLUSION

Studies were conducted on stationary diesel engine exhaust using plasma, plasma assisted catalyst and adsorbent hybrid processes. The major inferences drawn from this work are:

1. The NO_x removal performance of a plasma process is significantly affected by the exhaust flow rate. A double step/multistep plasma treatment seems to be a good option particular at large flow rates. This aspect becomes more relevant in a practical situation where the flow rate of the exhaust to be treated is significantly high.
2. The importance of aldehyde for the catalytic activity is confirmed both in the case of red mud catalyst and activated alumina catalyst. The aldehyde present in the diesel exhaust is just sufficient to activate red mud catalyst but not activated alumina catalyst.
3. The importance of by product formation of plasma process is seen in the case of plasma assisted catalyst hybrid process. The aldehydes formed in plasma assists catalysis. This is confirmed in the case of activated alumina which works as catalyst when it is assisted by plasma.
4. Plasma assisted red mud catalyst process is not much affected by the nature of exhaust. This is important in a practical situation where both plasma and catalyst can be made to treat filtered exhaust.
5. In single step plasma assisted catalyst hybrid process the type of catalyst plays an important role in NO_x removal while in double step plasma assisted catalyst hybrid process the type of catalyst has a little effect on the NO_x removal.
6. In single step plasma assisted adsorbent hybrid process the type of adsorbent plays an important role in NO_x removal while in double step plasma assisted adsorbent hybrid process the type of adsorbent has a little effect on the NO_x removal.

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