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 (NOx, 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 (NOx, 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 NOx, 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.**

Key words: *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 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 [11-25]. 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 (NOx, 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 (NOx, 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 NOx, 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. 1a and 1b. The fig.1a. represents the schematic for single step plasma based technique while fig.1b. represents the schematic for double step plasma based techniques.

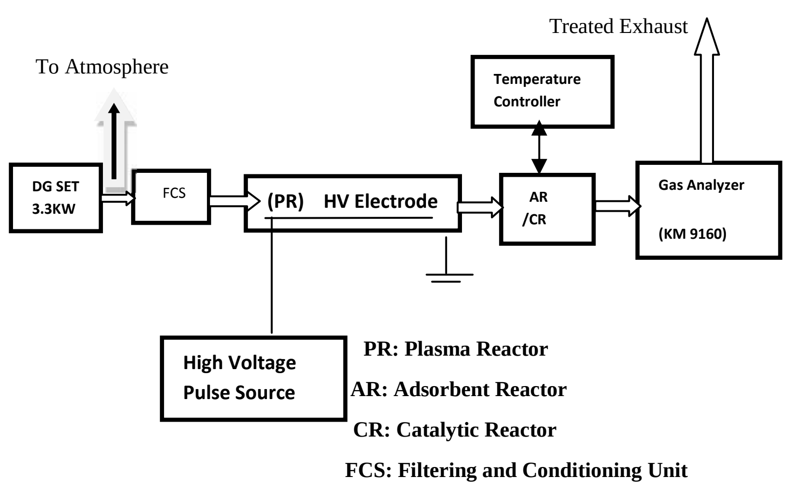

Fig.1a. Diesel engine exhausts treatment using single step plasma/plasma hybrid technique

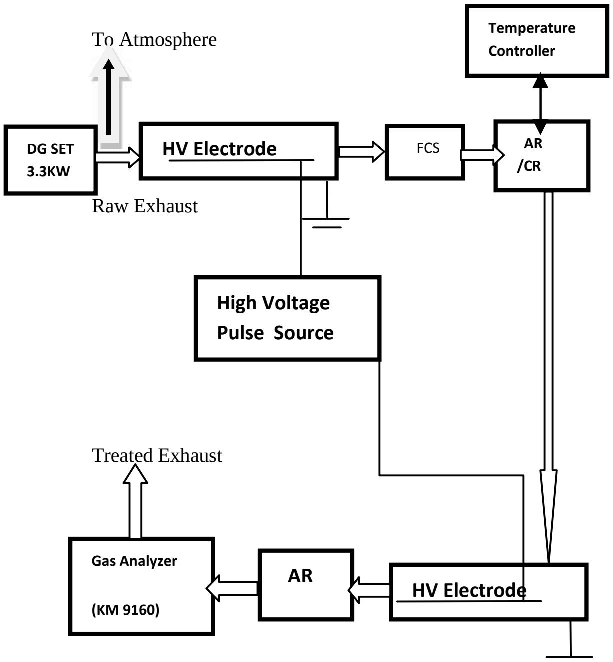

Fig. 1b. Diesel engine exhausts treatment using double step plasma/plasma hybrid technique.

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 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 30cm. 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 ^{0}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 NOx, 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, CO_2, NO, NO_2, NOx, O2and aldehydes** were measured. Table I shows the typical concentrations of the pollutants under 0% load and 27.27% load conditions.

Table.I
Initial Concentration of pollutents/components present in diesel engine exhaust

Main Pollutants	0 % Load(No load)	27.27% Load
H_2O	1.0% Vol	1.0%Vol
CO_2	0.10%	0.60%
CO	323ppm	229ppm
NO	110ppm	257ppm
NO_2	30ppm	69ppm
NOx	140ppm	326ppm
Aldehydes	50ppm	80ppm
O_2	20.70%	20.50%

In Table I, NO_x means sum of concentrations of NO and NO_2. 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.

(A) Plasma process

In this section both single step and double step plasma processes were discussed. Fig.2 shows the variation of NOx removal efficiency for different pulse voltages at various exhaust flow rates under no load in case of single step plasma process. The flow rate considerably affects the NOx 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 NOx removal is shown in fig.3.It is seen that for a given pulse voltage the NOx removal is not much affected by the flow rate.This could be attributed to the increased exhaust resident time in case of double step plasma process. This result is important while configuring the plasma reactors to handle large flow rates in practical situations.

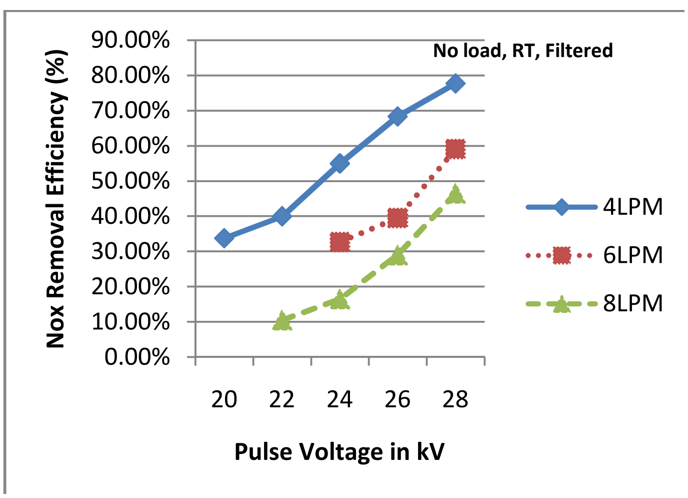

Fig.2. Effect of Flow rate on Single step plasma

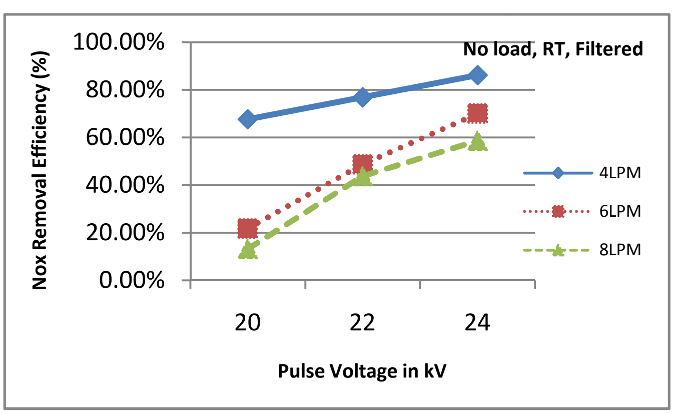

Fig.3. Effect of Flow rate on Double step plasma

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

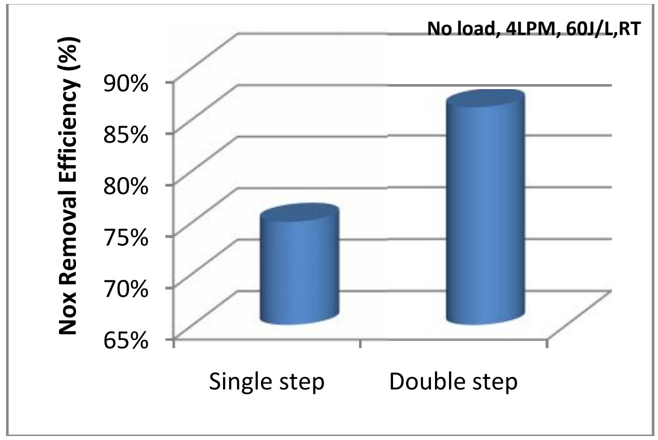

Fig.4. Comparison of single step and double step plasma

(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 NOx removal increases with temperature for red mud [26], while it decreases with temperature for activated alumina [27]. 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 Fe_2O_3, Al_2O_3, SiO_2, TiO_2, Na_2O, CaO, MgO and a number of minor constituents like K, Cr, V, Ni, Cu, Mn, Zn etc. Generally ferric oxide (Fe_2O_3) 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 m^2/g. Red mud has a fine particle size distribution with 90% by volume below size of 75 micron, and high surface area.

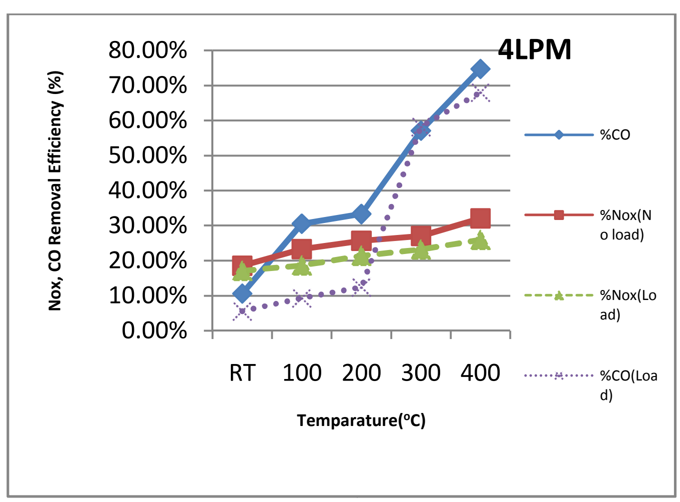

Fig.5. Effect of load on Nox & CO in Red Mud Catalyst removal

The fig.5 shows the effect of load on the NOx and CO removal by red mud catalyst for different temperatures. Various reaction pathways have been discussed for NOx and CO removal [26]. However the exhaust concentration is substantially high at load condition which reduces the activity of the catalyst in turn resulting in reduced NOx and CO removal under load condition.

The fig.6 gives the effect of flow rate on NOx and CO removal in red mud catalyst. It is seen that at high flow rates, both NOx and CO removal decreases and further the effect of flow rate is more pronounced with regard to CO removal than NOx removal. This can be due to reduced activity of the reactions involving CO[26]

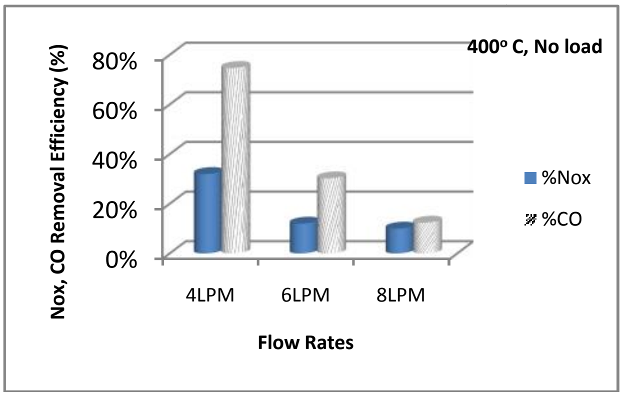

Fig.6 Effect of flow rate on Nox & CO removal in Red Mud Catalyst

(C) Plasma Assisted Catalytic 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

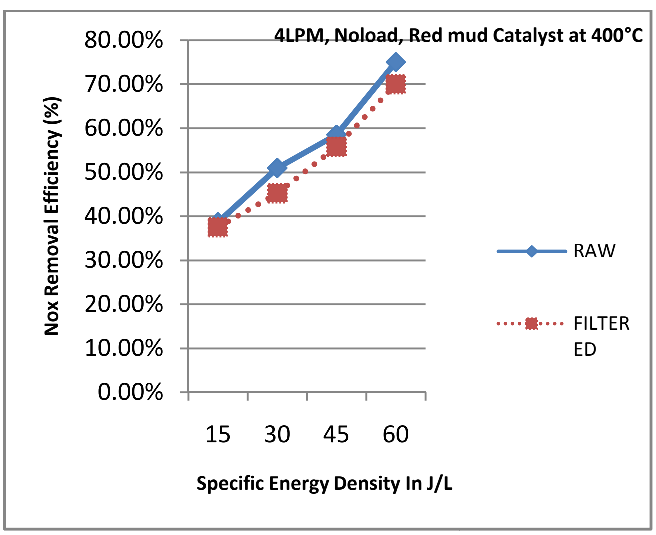

Fig.7.Effect of exhaust nature on single step plasma assisted catalytic process

The superior NOx removal performance of plasma treating raw exhaust has been well established [28].However when plasma is associated with red mud catalyst the effect of exhaust composition on NOx removal becomes significantly small as seen in the fig.7. In this fig for a given energy density the NOx removal is almost the same for filtered and raw exhaust. This can be explained as below:

When plasma assisted catalyst process treats raw exhaust the exhaust entering the catalyst contains less concentration of aldehydes which makes the catalyst less active. However when the plasma assisted catalysts process treats filtered exhaust, exhaust entering the catalyst contains more aldehyde which makes the catalyst more active [28]. Thus the exhaust composition has little effect on the NOx removal performance of plasma assisted catalyst process.

The fig.8 gives comparison between single step and double step plasma assisted catalyst process with regard to NOx 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 case of double step hybrid process the type of catalyst has a little effect on the NOx removal which can be due to a major contribution from the plasma process in the double step hybrid configuration.

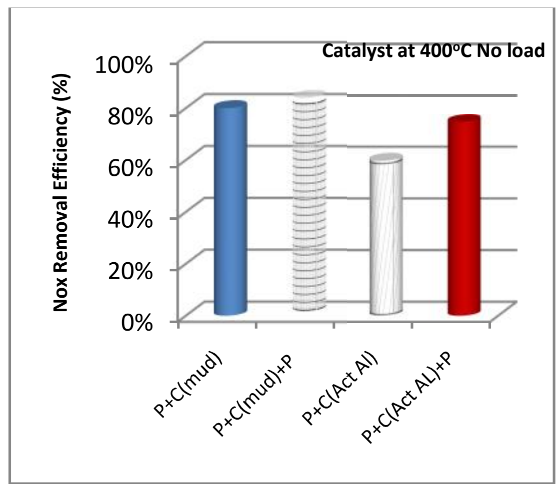

Fig.8. Comparison between single step and double step plasma assisted catalytic process

(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 [29]. The fig.9 gives comparison between single step and double step plasma associated adsorbent process with regard to NOx removal. In single step the hybrid process with MS13X as adsorbent performs better compared to activated alumina as adsorbent which can be attributed to better adsorbent action of MS13X [27,29]. However in case of double step hybrid process the type of adsorbent has a little effect on the NOx removal which can be due to a major contribution from the plasma process in the double step hybrid process.

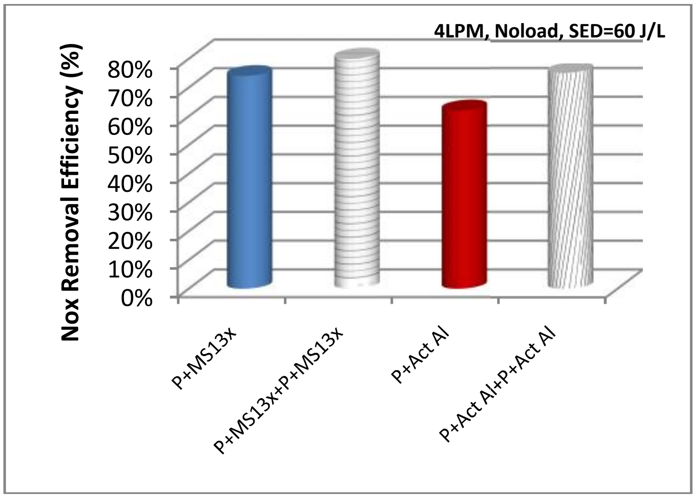

Fig.9 Comparison of single step and double step plasma assisted adsorbent process

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

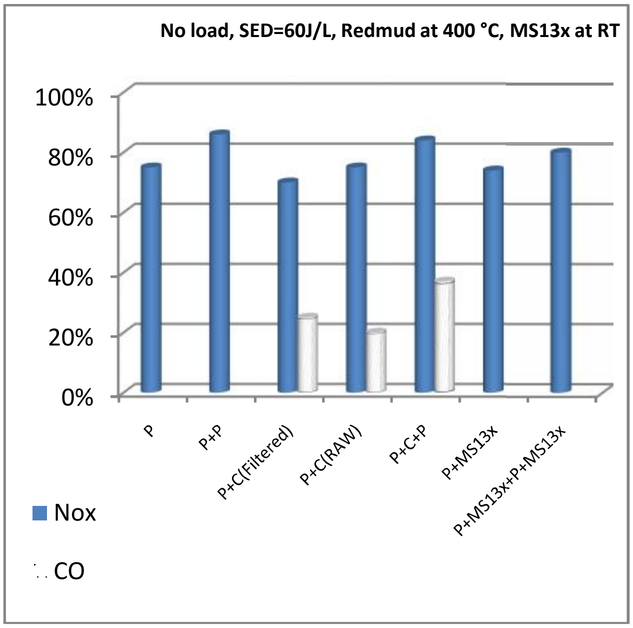

Figure. 10 Performance of different types of techniques for removal of Nox & CO

P: Single step Plasma reactor treating filtered exhaust, 0% load, room temperature
P+P: Double step Plasma reactor treating filtered exhaust, 0% load, room temperature
P+C (filtered) : Single step plasma assisted red mud catalyst reactor 0 % load, 400ºC
P+C(Raw): Single step plasma assisted red mud catalyst reactor 0 % load, 400ºC
P+C+P: Double step plasma- red mud catalyst reactor 0 % load, 400ºC
P+MS13X: Single step plasma assisted adsorbent hybrid technique, 0 % load, Room Temperature
P+MS13X+P+MS13X: Double step plasma assisted adsorbent hybrid technique, 0 % load, Room Temperature

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 NOx 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 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 case of plasma assisted catalyst hybrid process. The aldehydes formed in plasma assists catalysis. This is confirmed in 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 NOx removal while in double step plasma assisted catalyst hybrid process the type of catalyst has a little effect on the NOx removal

6. In single step plasma assisted adsorbent hybrid process the type of adsorbent plays an important role in NOx removal while in double step plasma assisted adsorbent hybrid process the type of adsorbent has a little effect on the NOx removal

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