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Original Article

Exhaust gases depletion using non-thermal plasma (NTP)

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ABSTRACT

The exhaust gases are a set of noxious gases produced as a result of combustion of fuel. They are the major component of environmental pollution and have deleterious effects on the humans' health. These gases expand gradually in the environment with an increasing demand for fuel consumption. An effective and precise technology is required to control the concentration of these gases. This paper discusses important modifications in the non-thermal plasma (NTP) to enhance depletion of the exhaust gases. The effectiveness of the procedure improves with the use of appropriate “filter”, keeping an adequate distance between the two corona plates, and a suitable gap between the electrodes and barrier. The adsorption of NTP is enhanced by increasing the RPM (revolution per minute) of the exhaust fans. It is observed that with the above modifications, the concentrations of CO, CO_2, HC, and NO_x are reduced by more than 95%, and a small amount of oxygen is also expected to form during the process.

1. Introduction

The plasma is a partial or fully ionized gas formed of different particles i.e. the electrons, atoms, ions, and molecules (Chang et al., 1991). The temperature of individual gases contributes to the formation of plasma. When all the components of a gas are in thermal equilibrium, it is called thermal plasma (Oda et al., 1993; Yamamoto et al., 1992). In the case of non-thermal plasma (NTP), the individual components do not have a similar temperature, i.e. they are not in thermal equilibrium. NTP is also known as a “non-equilibrium plasma”. The electrons have a small mass and can be mobilized by an external electric field. The electrons' temperature usually ranges from 10,000–250,000 K (Oda et al., 1993). The electrons can be easily accelerated to gain a high energy and produce free radicals from the parent molecules. The free radicals are reactive species, and actively bind with the environmental pollutants to form decomposition products (Penetrante and Schultheis, 1993). However, the harmful pollutants are scattered in a large volume of air, and the interaction of the free radicals with the pollutants is usually quite negligible.

The growing human technology makes a considerable use of fuel-based energy. The industrial machinery, aircraft's' engines, generators, and vehicles derived energy from the combustion of fuel. Energy is mainly derived from the fossil fuels, which includes solids, liquids, and gaseous fuels (Okumoto et al., 1997). The combustion of fuel in the engines produces a combination of heat and exhaust gases. The exhaust gases are an important component of the environmental pollution (Penetrante and Schultheis, 1993). The toxic gases in the environment can prove harmful to the human health, especially the lungs. Exposure and Inhalation of the harmful gases produce destruction in the airways and may impair their function. These noxious gases include Sox, No_x, Co, and CO_2. The emission of toxic volatile organic compounds (VOC) into the environment is also a significant problem (Shultz and Wulf, 1940; Tokunaga and Suzuki, 1984). Some of the VOCs are quite noxious and prove harmful to the human health and may also compromise the protective functions of the ozone. The significance of the problem has led to the development of a variety of technologies aimed at the control of the noxious gases. This include, Denitrogenation for the consumption of No_x (Chang et al., 1991; Yamamoto et al., 1992) Integrated Gasification Combined Cycles (IGCC), and non-thermal plasma (NTP) are valuable modalities used for the elimination of the harmful environmental gases.

The non-thermal plasma effectively reacts to the toxic pollutants in air, decompose them to non-toxic compounds and reduce their concentration. The researchers are interested in performing

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non-thermal plasma processing at the normal atmospheric temperature. A recent work suggested that the non-thermal plasma is quite effective in lowering the concentrations of toxic air pollutants.

Two important methods of cleaning the contaminated gas are found very useful in decreasing the energy consumption. One of them is a discharge method that uses a reactor's configuration and power supply. The parameters include frequency and waveform of the voltage (Oda et al., 2001; Belova et al., 1978). The second method uses catalysts for the VOC's decomposition or the removal of NO_X (Kim et al., 1999; Hoard et al., 2001; Shimizu and Oda, 1999; Oda et al., 2002). Some of the researchers have made a simulation of the non-thermal plasma. However, the estimated performance of the plasma processing is not yet clearly defined. It is also important to perform non-thermal plasma processing at the atmospheric pressure. It will be a new paradigm in technology. The analysis of the toxic or nontoxic materials produced in the plasma processing will improve the technique further. The NTP is a preferred method because it is more commercial and has a better efficiency. The plasma chemical processes promote effective oxidation with enhanced molecular dissociation and produce free radicals to accelerate the decomposition of toxic gases. Therefore the plasma chemical processes have many applications (Li et al., 2001; Yamada et al., 2002; Feng et al., 1998; Kawasaki et al., 2001). The impact of electrons provides the minimum amount of energy to break the bonds and destroy the Volatile Organic Compounds (VOC's).

The plasma processing triggered by the Corona discharge give oxide ions, molecular dissociation and produce free radicals for a rapid removal of toxic gases. In a non-thermal plasma processing the electrons' temperature ($T \gg 1$ eV) is quite higher than the temperature of the gas that includes the rotational and vibrational energy of the molecules ($1E = 0.1$ eV). It is known as an energetic electrons' process. The energetic electrons produce molecular excitation, dissociation, ionization and attaches to the lower energy electrons at the discharge area to form negative ions (Li et al., 2001). Secondary plasma reactions begin in the downstream afterglow discharge region between the dissociated molecules, radicals and ions. Aerosol particles are generated in the downstream afterglow discharge region, formed by the anions, growth of the cluster molecules and ions. Chemical reactions also take place on the surface of aerosol particles and depend on the composition of pollutant gas. Therefore, the decomposition of pollutants to nontoxic products in NTP involves the electron reactions, radical reactions, ion reactions, generation of aerosols and surface reactions.

This article will discuss the technique of increasing the efficiency of exhaust gases removal by the non-thermal plasma. The effectiveness of decomposition of pollutant gases like Sox, NO_X, HC, CO, O_2 and CO_2 were analyzed by changing the flow rates of gases and applying a filter to the NTP chamber. The flow rates of the exhaust gases can be changed by varying the RPM of the exhaust fans at the outlet of the corona discharge plates.

2. Experimental apparatus and technique

2.1. Experimental setup

Fig. 1 shows a detail of the experimental setup. The commercial gasoline was used in the engine to generate exhaust gases. 1.8L engine size with the power output of 140 horse power. The engine was operated in a steady state condition. The exhaust gases produced were then mixed with the fresh air at 1:5 and directed into the exhaust duct where the flow rates can also be analyzed. The analysis of the exhaust duct was carried out for the known exhaust gases like NO_X, HC, CO, O_2 and CO_2. The gases subsequently pass through the filter first and then the NTP reactor where the temperature and pressure are kept constant. HEPA filters, as defined by the United States Department of Energy (DOE) standard, this removes at least 99.97% of airborne particles 0.3 micrometers (μm) in diameter. The filter's minimal resistance to airflow, or pressure drop, is usually specified around 300Pa (0.044 psi) at its nominal flow rate. The pressure drop can be overcome through the exhaust fans. The voltage supply to the NTP chamber was also kept constant. Pulsed voltages with a frequency of 15 kHz with adjustable amplitudes (0–5) kV were applied across the electrodes to produce NTP power is supplied by the AC HV power supply. Power supply can be obtained by the transformer with a power amplifier (model 4510, NF Corporation, 5 kVA). The peak voltage that transformer can give is 20 kV. The NO_X and HC were analyzed with the help of particulate matter analyzer, while the CO, O_2 and CO_2 were analyzed by the auto-logic analyzer. As fresh air is added during the process, therefore, the relative humidity was also analyzed because it affects the NTP adsorption process. The flow rates of gases through the NTP can be changed by changing the RPM of the exhaust fans as shown in the figure. The numerical data was directly recorded with the help of Microsoft Excel and incorporated

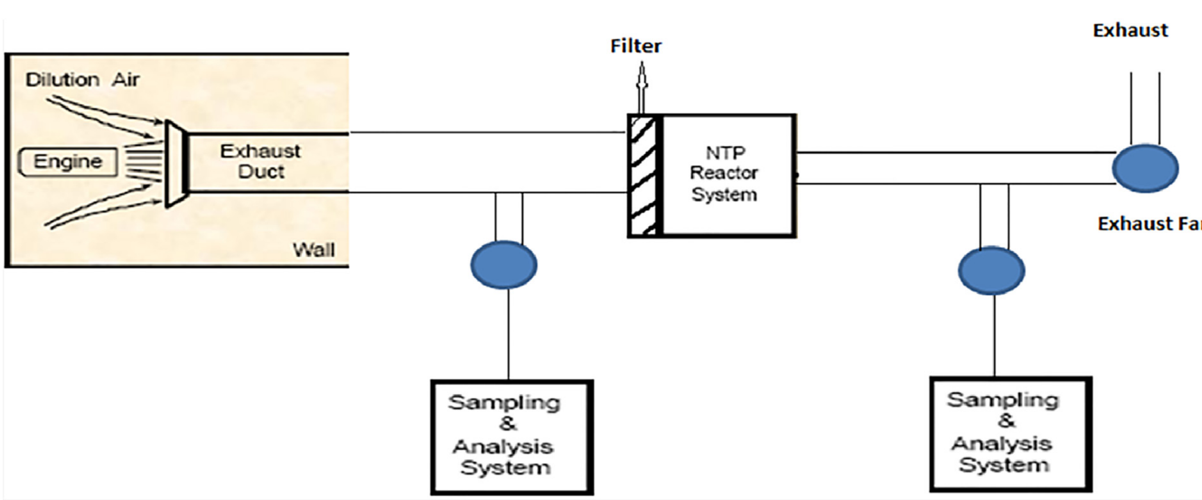

Fig. 1. Schematic diagram of the experimental setup.

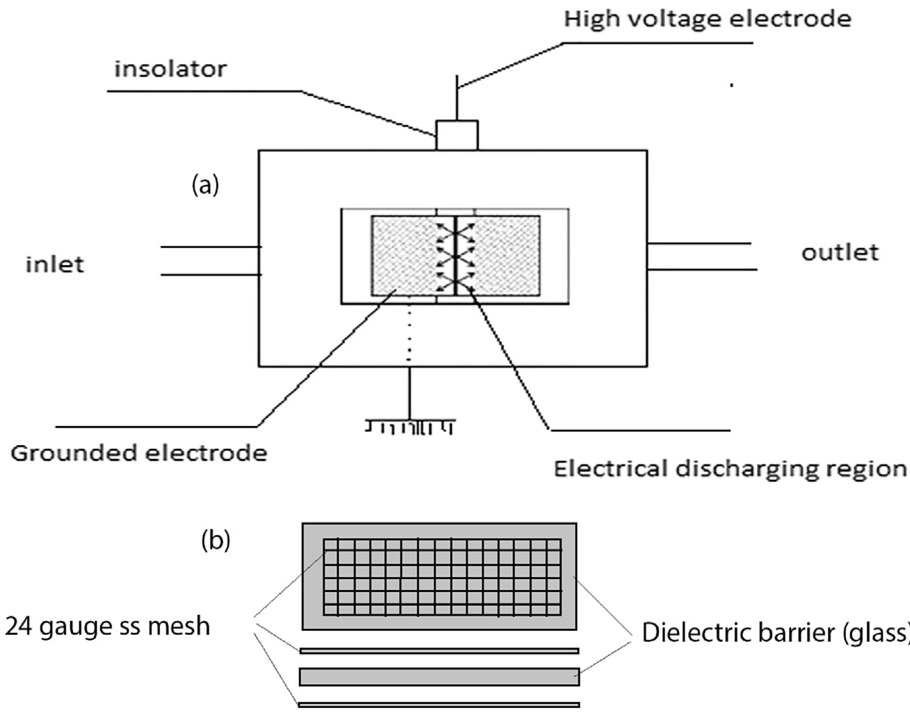

Fig. 2. Plasma reactor's sketch out side view (a) inside view (b).

in the auto-logic and particulates analyzers. AutoLogic Gas Analyzers are available in 4 or 5 gas configurations. 4 gas models measure HC, CO, CO_2 and O_2. 5 gas models include NO_x measurement. The analyzer uses a heavy duty non-dispersive infrared (NDIR) gas bench for measurement of HC, CO and CO_2. Electrochemical sensors measure O_2 and NO_x. Operating Temperature is 0°C–120 °C, Altitude: −300m to 2500m, Vibration: 1.5 G sinusoidal 5–1000 Hz, Shock: 1.22 m drop to concrete floor (gas analyzer), Response time: 0–90%, 8 s.

2.2. Plasma reactor

In Fig. 2 the plasma reactor consists of a rod type stainless steel electrode measuring 2 mm in diameter and a plate type electrode with a piece of glass placed on the surface of the plate which serves as a dielectric barrier. The effective discharge length was 90 mm and the discharge gap was 5 mm. Pulsed voltages with the frequency of 15 kHz and adjustable amplitudes of (0–5) kV were applied to the electrodes to form non-thermal plasma. A wattmeter connected to the low voltage side of a step-up transformer measured the input power (Willey, 1937; Belova et al.,1978). The wattmeter reading is subtracted from the transformer loss to obtain the input power of the dielectric discharge barrier. Power is supplied by the AC HV power supply. Power supply can be obtained by the transformer with a power amplifier (model 4510, NF Corporation, 5 kVA). The peak voltage that transformer can give is 20 kV.

Table 1
The concentration of gas components at the inlet.

Parameters	Concentration
HC (ppm)	10
NO_x (ppm)	116
CO_2%	3.32
CO%	0.5
O_2%	17.03

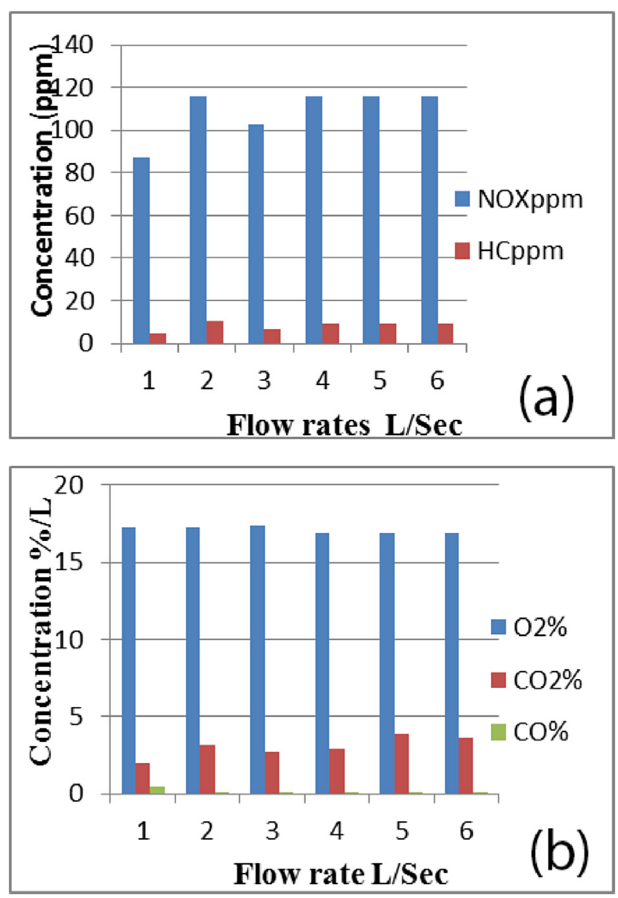

Fig. 3. Results of exhaust gases at different flow rates, (a) Particulates matters concentration (ppm) on different flow rates, (b) CO_2, CO and O_2 concentration (%/L) at different flow rates.

3. Results and discussion

3.1. Exhaust gases at the inlet

Table 1 shows the concentrations of NO, HC, O_2, CO, and CO_2 in the untreated exhaust gas produced when the gasoline engine was running in a steady state with an air-fuel ratio of 1.15. Fig. 3 shows the spectrum of the particulate matter's concentration of the exhaust gas and that of the outdoor air for comparison. The dilution ratio of 1:5 was used in the experiment and the flow rates of exhaust gases were changed from 1 L/sec to 6 L/sec.

Fig. 4(a) shows a relationship between the oxygen concentration in the exhaust gas at different flow rates, and the time interval from the beginning of the process. It can be appreciated that when the flow rate of the gas is increased from 1 L/sec to 6 L/sec, then the concentration of mixed oxygen is also increased by a factor of 2%. The increased content of O_2 in the exhaust gas is maintained for almost six hours as shown in the figure. After six hours the concentration of O_2 in the exhaust gas falls to the baseline. The highest concentration of O_2 in the exhaust gas was observed at a flow rate of 1 L/sec.

Fig. 4(b) shows the concentrations of CO_2 in the exhaust gas before treatment and after passing through the reactor's chamber at a particular flow rate. When the gasoline engine produces the exhaust gas and unless passed through the NTP's reactor, the CO_2 content increases with the passage of time as shown by the upper line in Fig. 2(b). However, when the flow rate of the exhaust gases increased through the NTP's chamber, the content of CO_2 began to fall rapidly with the passage of time. During the process, it was observed that the concentration of CO_2 decreased more rapidly when the gas' flow rate was 1 L/sec.

Fig. 4(c) shows the concentrations of CO in the exhaust gas before treatment and after being passed through the plasma reactor at a particular flow rate. The fall in concentration of CO is more efficient i.e. more than 98% when the exhaust gas is passing through the reactor's chamber at 1 L/sec. The removal of CO was not optimum at other flow rates.

Fig. 5 shows the effect of exhaust gas' flow rate on the removal of NO_x and HC at room temperature and keeping the energy density at 30 J/L. There is a significant fall in the concentration of NO_x with increasing flow rate of the exhaust gas, as shown in Fig. 3(a). As shown in Fig. 5(b), accelerating the exhaust gas by increasing the flow rate through the reactor decreases the concentration of both the NO_x and the HC. The efficiency of removal of the NO_x and HC becomes lower for the higher flow rates because the individual molecules do not have ample time to interact and decompose in the reactor.

3.2. The effect of temperature and energy density on the removal of exhaust gas

Fig. 6 shows the effectiveness of NTP in the removal of PM, HC, CO, and CO_2 at seven different ambient temperatures. During the experiment, the energy density of 40 J/L and the flow rate of 2 L/sec were maintained. Fig. 6(a) indicates that the removal of noxious gases and hence the effectiveness of plasma reactor are both temperature dependent. A higher ambient temperature reduces the heat loss of plasma and maintains a stable state of NTP. The bonds

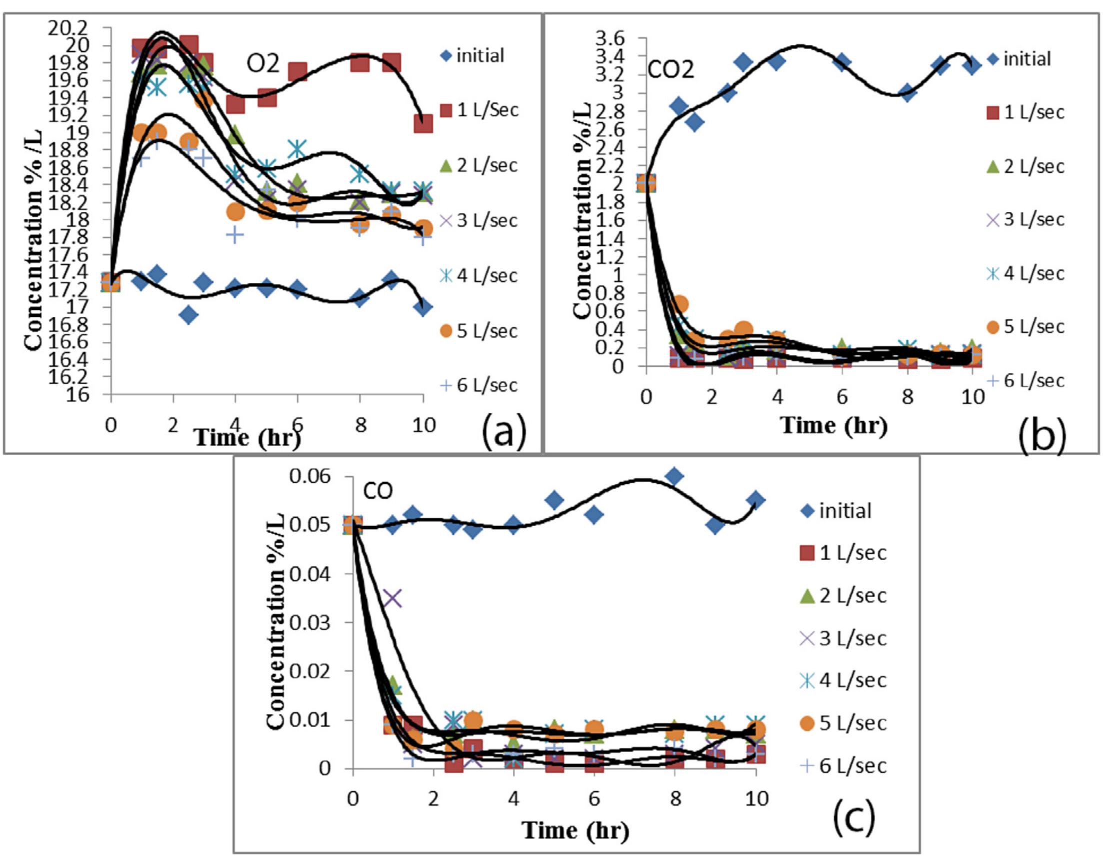

Fig. 4. Effect of flow rates on O_2, CO_2, CO concentration. The temperature was kept on 50° C and energy density at 30 J/L. (a) Oxygen concentration %/L on different flow rates, (b) CO_2 concentration %/L on different flow rates, (c) CO concentration %/L on different flow rates.

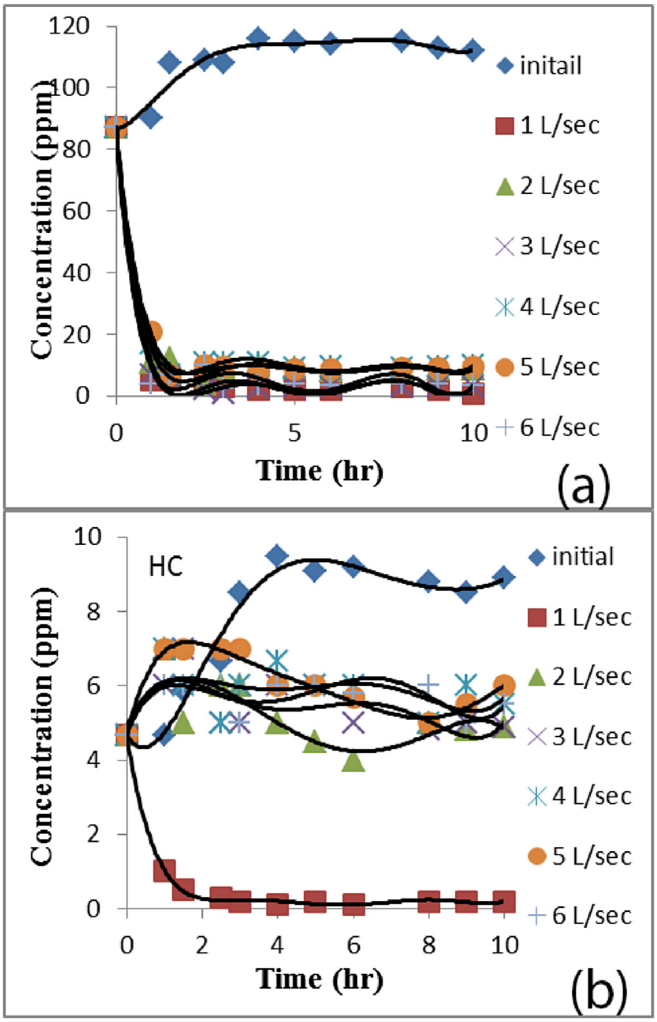

Fig. 5. Effect of flow rates on NO_x and HC concentration. The temperature was kept on 50° C and energy density at 30 J/L. (a) NO_x concentration %/L on different flow rates, (b) HC concentration %/L on different flow rates.

strengthening the VOCs' molecules are completely oxidized and readily dissociated at a higher ambient temperature in the plasma.

Fig. 6(b) shows the effectiveness of non-thermal plasma in the depletion of exhaust gases at room temperature when the flow rate was 2 L/sec. The decomposition of exhaust gases was more effective in the upper part of the lower range energy densities. The concentration of gas decreases when the density increased from 30 to 45 J/L.

The performance of NTP reactor depends on the presence of increased number of free radicals such as O, O_2, O_3, and nitrogen. The electrons are accelerated by an external electric field. They make a high energy impact with the component gases i.e. nitrogen, oxygen and carbon dioxide and produce free radicals. The number of free radicals produced during the process depends on the strength of electric field or E/N, where E is the strength of electric field and N is the density of gas. The electric field and energy density are related to each other.

Therefore, the production of free radicals increases by increasing the energy density. However, the benefit is limited to a certain threshold level of energy density and exceeding the limit may actually decrease the number of free radicals. A decrease in the number of free radicals results from a change in discharging condition. The color of the discharging region changes from violet to salmon pink when the energy density exceeds the threshold. This observation suggests that local arcs are formed due to the high strength of electric field, reducing its strength and as a result, a decreased amount of electric field's energy is delivered to the exhaust gases. The production of soot can be suppressed by oxidation of the soot's particles in a high-temperature (Okumoto et al., 1997). The oxidation at a higher temperature burns the smaller particles faster than the large size particles (Ohisa et al., 1999). The proposed chemical reactions (TMikoviny et al., 2004; Seung et al., 2010) are given below:

1) $CO_2 + e \rightarrow CO + O + e$
2) $CO + e \rightarrow C + O + e$
3) $3CO_2 + e \rightarrow 3CO + O_3$
4) $O + O \rightarrow O_2$
5) $NO \rightarrow N + O$
6) $NO_2 \rightarrow N + O + O$

4. Conclusion

Abundant amount of energy derived from the fuel combustion releases toxic gases in the global atmosphere and their effective removal becomes an essential part of the growing scientific research. There are many important methods to lower the concentration of toxic gases and protect the environment from their harmful effects, however, the yield of NTP is far better. The processing of NTP depends on the generation of high-speed electrons by an external electric field. The accelerated electrons ionize the component exhaust gases, generate free radicals, initiate a chain of

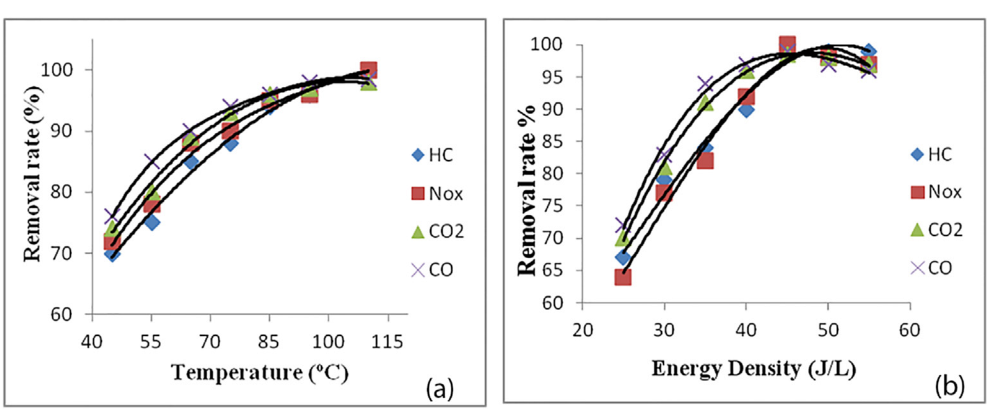

Fig. 6. Effects of Temperature and Energy density on HC, NO_x, CO_2 and CO. Flow rate was kept at 2 L/sec, (a) Effect of Temperature on HC, NO_x, CO_2, CO removal rate. (b) Effect of Energy density on HC, NO_x, CO_2, CO removal rate.

chemical reactions, and decompose the noxious gases into non-toxic products. We have attempted to increase the rate of decomposition by increasing the flow rates of the exhaust gases through the NTP reactor. We concluded from our experimental work that a rapid fall in the concentration of HC, NO_x, CO and CO_2 occurs at a flow rate of 1 L/sec. The removal rate or efficiency of NTP's reactor decreases when the flow rate exceeds 1 L/sec because the time of contact of the exhaust gases decreases in the NTP's reactor. A greater benefit of the non-thermal plasma is observed by increasing the ambient temperature of the exhaust gases. The removal efficiency also depends on the energy input and a better performance of NTP's reactor was observed at 45 J/L.

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