NO\textsubscript{x} Reduction by Atmospheric Pressure Dielectric Barrier Discharge

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ABSTRACT

Advanced oxidative and reductive reactions based on non-thermal plasma have been used to reduce NO\textsubscript{x} gases emitted from 4kW free load diesel engine exhaust. Coaxial dielectric barrier discharge reactor derived by 50 Hz sinusoidal power supply (0-12kV) has been used. Different parameters including, discharge power, discharge length and applied voltage have been investigated experimentally. The discharge cell has been optimized to provide a maximum NO\textsubscript{x} removal percentage of about 73\% and a maximum removal efficiency of about 32 gm/kWh have been obtained.

INTRODUCTION

Dielectric barrier discharge is regarded as non equilibrium plasma since the energy is deposited into electrons rather than ions or molecules, leaving the gas mixture essentially at the temperature of the environment. High-energetic electrons react with the gas molecules producing reactive species (radicals, excited molecules) that can initiate and sustain oxidative and reductive chemical reactions suitable for environmental applications\cite{1,2}, mostly include fields of air pollutant control,\cite{3} wastewater and drinking water decontamination\cite{4}, VOC treatments and NO\textsubscript{x} reduction\cite{5,6}.

In dielectric barrier discharge (DBD) plasma, the discharge occurs in a number of individual tiny breakdown channels of nanosecond duration which are distributed uniformly in the discharge gap and referred as micro-discharges. Filaments can be characterized as weakly ionized plasma channels with properties resembling those of transient high-pressure glow discharges. In a micro-discharge a large fraction of the electron energy can be utilized for exciting atoms or molecules in the background gas, thus initiating chemical reactions \cite{7}.

Emissions of NO\textsubscript{x} from combustion engines are primarily in the form of NO generated as a function of air to fuel ratio \cite{8}. NO produces the same failure to absorb oxygen into the blood as carbon monoxide (CO). It is believed that NO\textsubscript{x} be a major contributor to acidic deposition of acid rain \cite{9}.

The aim of the present paper is to design and optimize atmospheric pressure dielectric barrier discharge (APDBD) plasma source which can be operated at low power to achieve maximum reduction of NO\textsubscript{x} emitted from 4 kW free load diesel engine exhaust without contribution of any chemical catalyst. Different parameters of the plasma source have been optimized for maximum removal efficiency, including discharge length, applied voltage and dissipated power

Experimental Setup

Schematic diagram of the co-axial DBD system is shown in figure1. The discharge cell consists of two coaxial electrodes separated by a Pyrex glass tube with inner diameter of 1.2 cm, outer diameter of 1.5 cm and length of 140 cm. Copper rod was used as inner electrode with diameter of 0.8 cm and 120 cm in length drilled at one of its ends to allow gas flow to the discharge zone. A graphite layer pasted on the outer
surface of the glass tube was used as the outer electrode with a variable length of (10 to 120 cm). The gap distance between the inner electrode and the inner surface of the glass tube was set at 2 mm. Step up high voltage transformer derived by a variac was used as power source to deliver sinusoidal variable applied voltage from 0-12 kV at 50 Hz.

The current and voltage waveforms during the discharge were recorded by 100 MHz two channel digital storage oscilloscope (Type HM1508) where one channel was connected to a potential divider (500:1) to measure the applied voltage on the discharge cell, the other channel was used to measures the discharge current by measuring the potential drop across a 100 ohm resistance (connected between the cell and ground). The 100 ohm resistance was also replaced by a capacitor of 10 nF that measure the charge accumulated on the discharge cell. The discharge power is calculated using the Lissajous figure following the method of Manley [10,11].

The source of NOx gas was a diesel exhaust emitted from a 4 kW diesel engine operated at no load. The treated gas was directed to multi gas analyzer (model Teknotest 488) at flow rate of 5 L/min to measure NOx concentration. The initial concentration of the NOx was kept constant at 200 ppt.

RESULTS AND DISCUSSION

The current and voltage waveforms of the coaxial AC DBD system are shown in Figure 2. It can be observed that the mode of discharge is filamentary mode due to the nanosecond current filaments which is observed in the current waveform.

![Fig.1 Schematic diagram of the Coaxial AC DBD](image)

![Fig.2: The current and voltage waveforms of the coplanar coaxial AC DBD system](image)
Figure 3 represents the discharge power in the coaxial DBD system as a function of the discharge length. The increase in the discharge power with the discharge length is attributed to the increase in the discharge volume which increases the dissipation of the power inside the discharge cell.

Figure 4 shows the variation of NOx removal percentage as a function of the applied voltage at different discharge lengths (from 15 to 120 cm). The NOx removal percentages can be seen to increase by increasing the applied voltage for all discharge lengths. The expected oxidation/reduction reactions of NOx initiated in the plasma reactor in the presence of the exhaust gas of diesel engine (a mixture of NO, hydrocarbons, N2 and O2 gases) are summarized as follows [1,5,6]:

\[
\begin{align*}
O_2 + e & \rightarrow O^+ + O + e \quad k = 1.1 \times 10^{-12} \text{ cm}^3/\text{s} \\
O_2 + O & \rightarrow O_3 \\
N_2 + e & \rightarrow N + N + e \quad k = 3 \times 10^{-16} \text{ cm}^3/\text{s}
\end{align*}
\]

According to equation (1), the kinetic energy of the electrons is consumed primarily into the major gas components, N2 and O2 which results in the production of N and O atoms through electron-impact dissociation reactions.

Consequently another expected reaction takes place to change NO2 into NO as follows [5,6]:

\[
\text{NO}_2 + O \rightarrow \text{NO} + \text{O}_2 \quad k = 10^{-13} \text{ cm}^3/\text{s}
\]

The NO reduced into N2 through the following reduction reaction [5,6]:

\[
\text{NO} + N \rightarrow N_2 + O \quad k = 2.1 \times 10^{-11} \text{ cm}^3/\text{s}
\]

Fig. (3): The discharge power as a function of the discharge length

Fig. (4): NOx removal percentage as a function of applied voltage at different discharge lengths
Figure 5 shows the NOx removal percentage as a function of the discharge length. The reduction of NOx by increasing the discharge length is mainly attributed to the increase of the residence time of the reactants in the plasma reactor.

The efficiency of NOx removal can be expressed as the ratio between the removed mass of the NOx and the input energy of the discharge system. In other words, the NOx removal efficiency can be expressed as the ratio of the removal rate of NOx to the consumed power through the discharge cell. Thus the removal efficiency (Eff) can be expressed as follows:

$$Eff = \frac{R_r}{P}$$

(4)

Where $R_r$ is the removal rate of NOx and $P$ is the power consumed during removal process. The removal rate of NOx is given by:

$$R_r = R\% \times F$$

(5)

Where $R\%$ is the NOx removal percentage and $F$ is the gas flow rate.

Figure 6 shows the efficiency of NOx removal in DBD coaxial system as a function of the discharge length. The efficiency can be seen to decrease from 35 to 5 gm/kWh as the discharge length increases from 10 to 120 cm. As the discharge length increases, the consumed power increases (see figure 3) which follow the observed decrease in efficiency.

In the view of the present conditions of the AC DBD coaxial system the optimum maximum removal percentage and efficiency are as follows:
- The maximum removal percentage is 73 % at efficiency of 5 gm/KWh.
- The maximum efficiency is 32 gm/KWh at removal percentage of 35 %.

![Figure 5: NOx removal percentage as a function of the discharge length in AC DBD coaxial system](image-url)
CONCLUSION

Coaxial dielectric barrier discharge plasma source driven by 50 Hz sinusoidal feeding power supply (0-12kV) has been used for NOx removal. The discharge cell has been optimized for maximum NOx removal rate and removal efficiency. Maximum NOx removal percentage of about 73% and maximum removal efficiency of about 32 gm/kWh have been obtained.

REFERENCES