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# Removal of main exhaust gases of vehicles by a double dielectric barrier discharge

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**Abstract.** Because the health effects and their contribution to climate change, the emissions of toxic gases are becoming more controlled. In order to improve the diminution of toxic gases to the atmosphere, several techniques have been developed; here it will be focus only to automotive emissions. This work deals about the treatment of toxic gases emitted from vehicles by a non-thermal plasma. Several tests were done in a 4-cylinder 2002/Z16SE motor to characterize the vehicle emissions. With these results gas mixture simulating the exhaust gases vehicles, was used in experiments at different conditions employing a double dielectric barrier reactor for their treatment. The removal efficiencies superior to 90% show the competence of the non-thermal plasma reactor to treat these gases. Experimental results are explained with the aid of a simple chemical model that suggests a possible mechanism of degradation of toxic gases. The plasma reactor employed could works at 12V supplied without difficulty by a vehicle battery.

## 1. Introduction

Pollutants like nitrogen oxide (NOx), particulate matter (PM), sulfur dioxide ($SO_2$), volatile organic compounds (VOCs) and carbon monoxide (CO) are mostly generated by vehicles. The situation becomes more difficult because the inexistence of catalytic converter in around 30% of the vehicle fleet [1] and because the poisoning of catalytic converters with sulfur still present in Mexican gasoline. In January 2012 the Mexican Center for Environmental Rights [2] reports 38,000 death produced by atmospheric pollution between 2001 and 2009.

European regulations [3,4] are now focused on reducing greenhouse gases, above all carbon dioxide ($CO_2$) and, subsequently, modifications in vehicles have been done in order to diminish it; particularly the consumption of diesel is enhanced. The diesel, successfully, diminishes $CO_2$,

nevertheless the NOx and PM emissions increase and the use of conventional catalytic converters is not adequate [5].

The problem of toxic gases emission persists and the use of alternative technologies becomes essential. One promising alternative is the use of non-thermal plasma as it can be depicted in next paragraph.

In the branch of automotive pollution control several reactor configurations have been proposed (i.e. cylindrical pulsed corona [6,7], packed pellet cylindrical configuration [8], dielectric discharge with flat electrodes [9]). All these configurations show a good removal of $NO_x$ and PM; the packed cylindrical configuration has a relative better efficiency certainly due to a major plasma-polluted gas interaction, however in this type of reactors the exhaust gas pressure diminishes when the gas collides with a barrier (pellets), this fact implies an augment in consumption of gasoline in vehicles.

In this work a double dielectric barrier in a cylindrical configuration (2DBD) reactor is proposed to reduce gaseous emission of vehicles. The double dielectric allows a better distribution of plasma in the reactor; therefore a higher residence time of toxic gas was expected without diminishing the gas exhaust pressure.

## 2. Basic concepts

The chemistry in plasmas in a non-thermodynamic equilibrium state or commonly named cold plasmas are mostly based on radicals reactions, obtained from electronic collisions. For the chemistry model in a double dielectric barrier reactor, a detailed work was done in [10]. In this case the kinetic model used to represent the exhaust gas treatment was divided in two phases.

During the first step, it is considered an energetic (500 Td [11]) and quasi instantaneously ($1\times10^{-8}$ s) streamer head is formed, with an initial electron density of 1 $cm^{-3}$ [12-14]. In this phase two mechanisms for active species formation were considered: dissociation by direct electron impact with gas molecules and quenching of the excited states such as $O(^1D)$ and $N_2(A)$. Some primary radicals ($OH^\bullet$, $O^\bullet$, $N^\bullet$) appear during this phase. The radicals production is supposed uniformly distributed within the whole volume. The formation of a streamer channel connected to the streamer head is considered as the second phase; where NOx, propane and $CO_2$ react with active species.

The reaction mechanisms of the NOx-$C_3H_8$ and $CO_2$ degradation are respectively showed in figure 1(a) and (b). These mechanisms takes into account thirty three species ($e^-$, $N_2$, $O_2$, $H_2O$, $O^\bullet$, $N^\bullet$, $OH^\bullet$, $H^\bullet$, $O_2(a^1\Delta g)$, $O(^1D)$, $N_2(A)$, $O^-_2$, $O^-$, $O^+_2$, $H^-$, $O_3$, $NO_2$, $NO_3$, $N_2O_5$, $N_2O$, $HNO_3$, $HNO_2$, $C_3H_8$, i-$C_3H_7$, $(CH_3)_2CHO_2$ , $(CH_3)_2CHO$, $CH_3C(O)CH_3$, $CO_2$, CO, $CO^{2-}_3$,COOH, HCOOH,C).

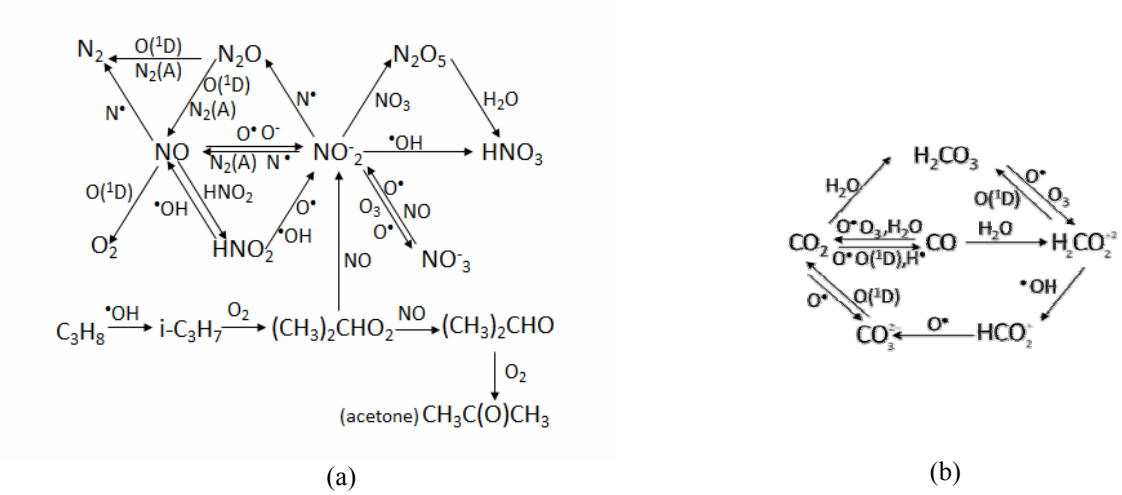



**Figure 1.** Chemical mechanism proposed (a) NOx-$C_3H_8$ conversion, (b) $CO_2$ conversion.

## 3. Experimental setup

The analysis of the gas exhaust in a 4-cylinder 2002/Z16SE motor (figure 2(a)) was realized under several conditions of acceleration (expressed in revolutions per minute- rpm) directly bounded with the opening of the butterfly valve and the weight transfer, denoted by the change in the load borne during acceleration. These conditions simulate different situations during vehicle driving, notably the acceleration and the increasing load during a hill climb, for example.

The gas exhaust analysis was realized with PG-250 and IR-FGA400XDS sensors that respectively detect NOx, $SO_2$, $CO_2$, CO, $O_2$ and Total Hydrocarbons. In relation to the quantification of propane, it cannot be effectuated; a qualitative analysis was only completed by employing Fourier transform infrared spectroscopy (FTIR) equipment.

Concerning the gas treatment by plasma, in figure 2(b) the experimental setup is schematized, it consists in a tube of Pyrex glass with an internal radius of 11.1 mm, a length of 75 mm and a thickness of 1.2 mm. A stainless steel concentric electrode of radius of 2.54 mm is set inside the Pyrex tube and a tube of alumina is wrapping it. A metallic mesh covering the Pyrex tube plays the role of the external electrode. The ideal mixture simulating the gas exhaust of the motor studied is continuously introduced into the 2DBD reactor at a gas flow of 3 liters per minute. An applied power ($P_A$) of less than 30W was applied to maintain the plasma. The outlet gases from the reactor were analyzed with the PG250 and a mass spectrometer RGA. An electrical diagnosis was also realized in order to calculate the consumed power (Pc) and power efficiency ($P_E$).

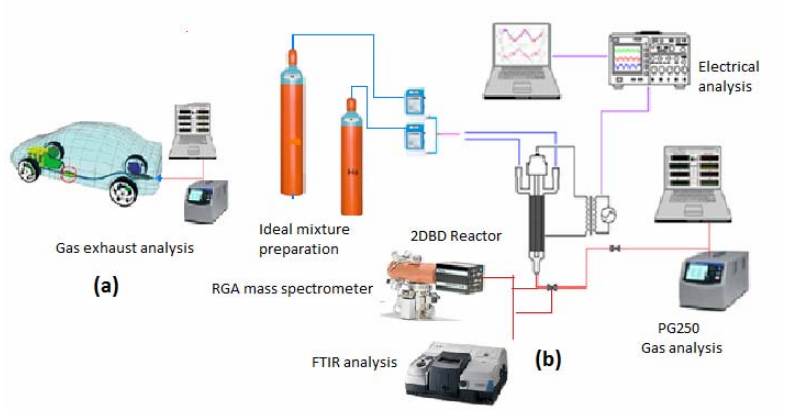



**Figure 2.** Experimental setup. (a) Exhaust gas characterization (b) Characterization of toxic gases treatment by a 2DBD.

The composition of the gas mixture to be treated with the plasma reactor is detailed in table 2. This composition was taken from results obtained during the analysis of the motor exhaust gases (reported in next section).

**Table 1.** Mixture of gas treated with plasma.

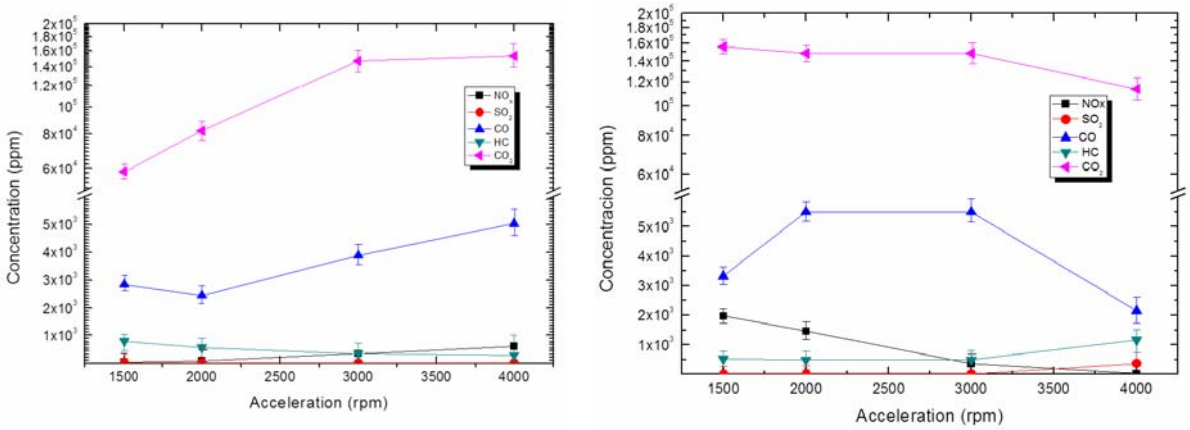
| Compound | Concentration |
|---|---|
| $C_3H_8$ | 1200ppm |
| CO | 4% |
| $CO_2$ | 12% |
| NO | 3000ppm |

## 4. Results

### *4.1. Motor emissions characterization*

Several driving conditions in the motor are reported in figure 3. Generally, an augmentation of noxious emissions is obtained with the increase of rpm. This could be explained by the fact that a higher number of rpm is the result of opening the butterfly valve and then, a higher air alimentation producing the oxidation of the gases due to higher temperatures and higher concentrations of oxygen.

In general, the concentration of NOx varies from 30 ppm to 2000 ppm, the CO augments from 2500 to 5000 ppm, the total hydrocarbons (HC) concentrations vary from 500ppm to 1200ppm and the rang covered by $CO_2$ concentrations is from 5%v.v to 15% v.v. Experimental errors fluctuates from 2 to 4.5%. All these concentrations are more important at higher rpm and at higher weight transfer.




(a) (b)

**Figure 3.** Motor conditions (a) without load, (b) with load.

When weight transfer is bigger (figure 3(b)) at 4000 rpm an unusual phenomena can be observed; the $CO_2$ and CO concentrations diminish, the oxidation of nitrogen and sulfur compounds to $NO_x$ and $SO_2$, seems to be favored. At higher temperatures the Zeldovich mechanism could be dominant in chemical reactions giving preference to the NOx formation [15]. For the $SO_2$ formation it is shown that the increase in temperature corresponds to a decrease of equilibrium constant values as result of reactions in direction of the reactants formation [16].

In both graphics presented in figure 3, the $SO_2$ concentration is not as much as 15ppm, except at 4000 rpm and higher weight transfer, where the concentration attains 372ppm. In order to simplify the experiences the $SO_2$ was not taken into account.

### *4.2. Plasma reactor to treat noxious emissions*

Several experimental conditions were tested; the better frequency of operation fluctuates at around 16 kHz. The applied power varied from 10 W (1 A, 10 V) to almost 30 W (1 A, 27 V).

A characteristic diagram of current and voltage in function of time is reported in figure 4. A relatively stable and continuous discharge with some microdischarges could be appreciated in the current curve. The discharge is a bit more stable than these found in a single dielectric barrier as was reported by Valdivia et al [17].

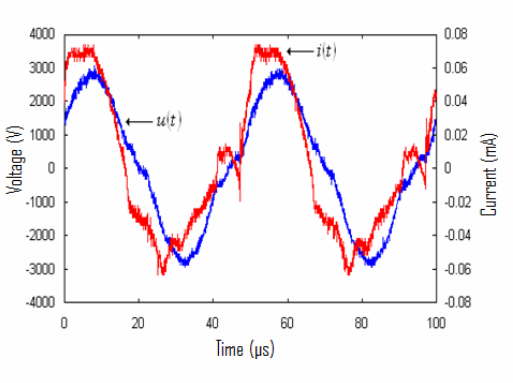



**Figure 4.** Voltage and current signals.

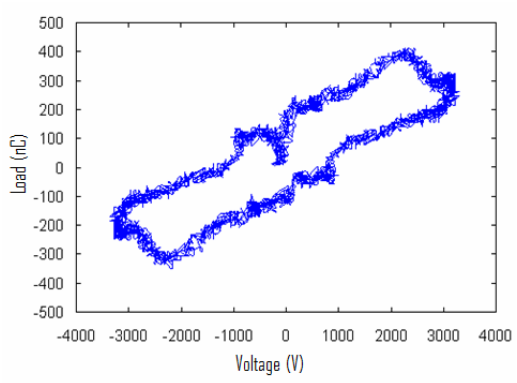



**Figure 5.** Lissajous diagram.

The Lissajous diagram depicted in figure 5 is representative of the dielectric barrier discharge that includes both the phase shift between the voltage and current signals and the accumulated charge in the dielectric of the reactor glass wall. This dielectric material prevents both the effect of electric arc and the microdischarges in the plasma. The electrical power $P_C$, was calculated; this oscillates between 7 and 23 W. After obtaining these values, and considering the power $P_A$, can finally achieve the efficiency $P_E$, as reported in table 2.

**Table 2.** Influence of electrical parameters into gas treatment

| Frequency (kHz) | $P_A$(W) | $P_C$ (W) | $P_E$ (W) | $NO_x$ removal (%) | CO removal (%) | $CO_2$ removal (%) |
|---|---|---|---|---|---|---|
| 16 | 10.30 | 8.18 | 79% | 94.54 | 43.99 | 30 |
| 16.03 | 10.40 | 9.43 | 83% | 96.9 | 15 | 25 |
| 16.8 | 27.26 | 23.7 | 86% | 96.24 | 37.64 | 15 |
| 16.82 | 20.85 | 17.95 | 86% | 98.03 | 15 | 30 |

From table 2, it can be observed that $NO_x$ removal is quite good (superior to 94%) at any condition, nevertheless the best removal was obtained at higher power efficiency.

For CO and $CO_2$ removal, the influence of the electrical conditions studied seems to be not conclusive; a possibly explanation of the oscillation in both concentrations is the competition for $O^{\bullet}$ radical as observed in figure 1(b).

The FTIR spectra obtained are reported in figure 6; here, when plasma is applied (figure 6(b)), a diminution of the propane can be qualitatively observed at around 3000 $cm^{-1}$. Another characteristic band that appears in the products at around 1700 $cm^{-1}$ is characteristic of nitric acid. The formation of acids is corroborated by a conductometer; a concentration of 20-40 ppm of $HNO_3$ was found.

From mass spectrometry study (figure 7), several compounds in products were formed after the plasma treatment; for example, in figure 7(b) the line situated at 17 uma, corresponds to $O^{-2}$, $O^{+}$ and $OH^{-}$. Line at 30uma depicts the $N_2$ and NO. It could be observed that acetone is also formed, reinforcing the mechanism schematized in figure 1(b) and also proposed by Dorai [18], however the propionaldehyde founded in [20] is not obtained at our experimental conditions, possibly because the concentrations of radicals $^{\bullet}OH$ are not enough.

## 5. Conclusions

The degradation of the chemical compound gas: NO, CO, $CO_2$, and $C_3H_8$ is successfully through chemical reactions and mainly free radicals like: $O^{\bullet}$, $NO^{\bullet}$ and $^{\bullet}OH$, this compounds are produced

basically through electronic collision with neutral molecule of water stream, oxygen and nitrogen. The degradation removal efficiency results for the automotive mixture depicts the feasibility to use in the future this technology of non-thermal plasma for the treatment of automotive toxic gas pollutants by taking care of remove the acids formed by using simply means, like activate charcoal.

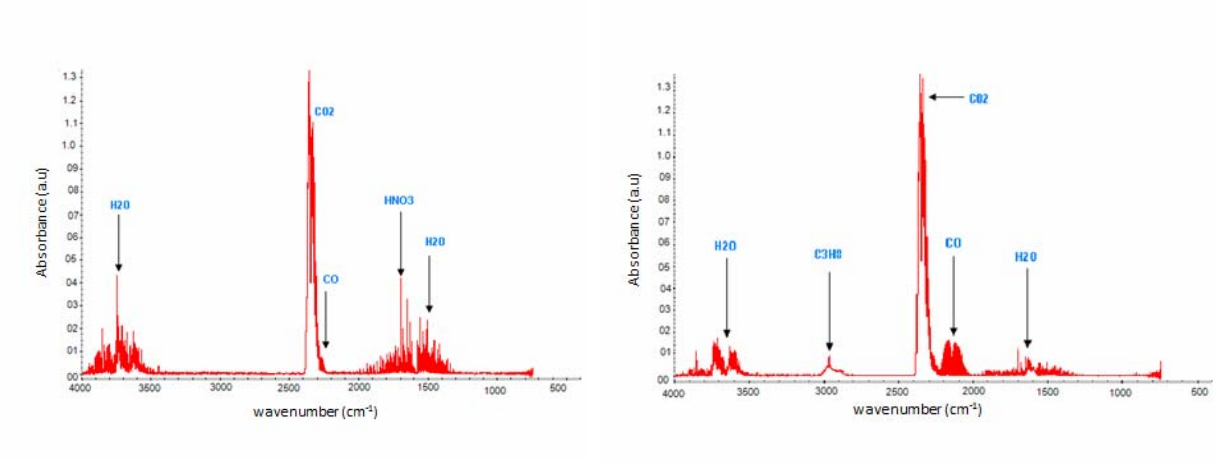
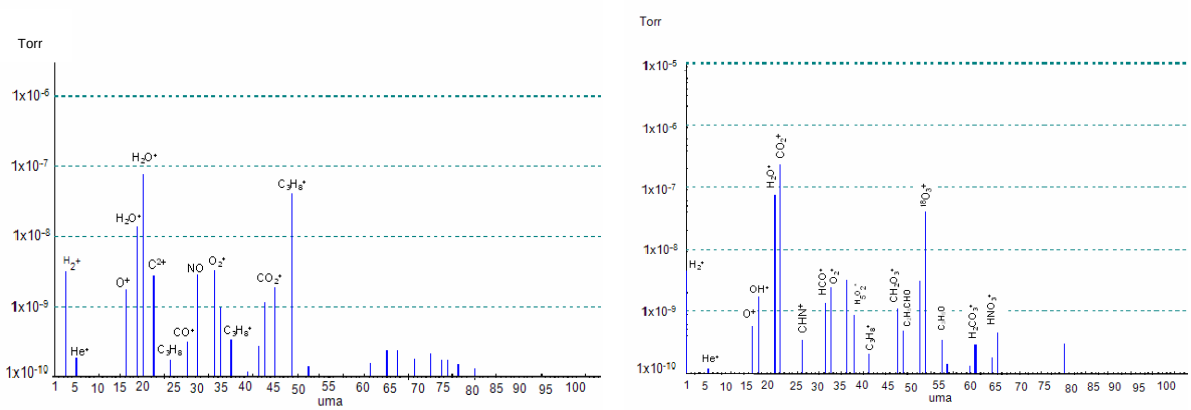



**Figure 6.** FTIR analysis of (a) non treated gas mixture and (b) after treatment products.




(a) (b)

**Figure 7.** Mass spectra of automotive emissions (a) without plasma treatment (b) with plasma treatment.

**Acknowledgments**
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