NOx Removal Enhancement by a *Jerks – and – Jumps* Type Electrode in a Dielectric Barrier Discharge

A. Mercado-Cabrera1*, E. León del Villar1, R. Valencia-Alvarado, R. López-Callejas12, S. R. Barocio1, R. Peña-Eguiluz1, A. Muñoz-Castro1, B. Jaramillo-Sierra2, A. de la Piedad-Beneitez2.

1 Instituto Nacional de Investigaciones Nucleares, Laboratorio de Física de Plasmas
AP 18-1027, 11801, México D. F.
2 Instituto Tecnológico de Toluca, AP 890, Toluca, México

*e-mail: amc@nuclear.inin.mx

Abstract: In this study, the electrode surface of a NOx removal treatment reactor has been modified in order to reduce its electric potential level and, at the same time, to increase its removal capacity by generating a cold plasma using a non-homogenous electric field on the electrode surface. This electric field has been achieved by means of a *jerks and jumps*-like electrode profile. The other electrode conserves the original flat form. Then, experiments on the removal of NOx were carried out in this 22.4 cm³ reactor. Concentrations of 30-80 μmol/mol of NOx in nitrogen were used with 1 SLPM flows. The exhaust gases were analysed as well as characterised by gas chromatography and mass spectrometry. Additional experiments were also carried out in a second reactor of the same reaction volume but where two conventional flat and parallel electrodes were used, in order to compare the results. The NO removal efficiency in the two flat electrode case approached 87% while ~98% in the *jerks and jumps* reactor.

1. Introduction
Recent successful applications of non thermal plasmas include, particularly, processes of gas flow decontamination. One of the main objectives of these techniques is avoiding that the gas temperature increase substantially [1] and that dangerous secondary compounds are not generated. Some non thermal plasmas are conditioned by the free electron generation at high energies by means of a dielectric barrier discharge [2]. These electrons interact with the background gas to generate reactive species, such as free radicals, which react in turn with compounds such as NOx [3, 4]. Unlike the oxidation process, in this modality a chemical reduction is used. In order to carry out the present work, a gas mixture has been treated in a newly designed reactor where the flat surface of one electrode has been modified in order to produce a controlled dielectric discharge. The removal efficiency results have been compared with those obtained from a traditional reactor of parallel electrodes with double dielectric.

2. Experimental Set-Up
A gas mixture of nitrogen oxide balanced with nitrogen gas was selected, with concentrations around 30, 60 and 80 μmol NO /mol. The total mixture flow was fixed at 1.0 LPM. The concentration during all the tests was measured with a Sensonic 2000 gas analyzer. The concentration was also gauged by
means of a 6890N network gas chromatographer coupled to a 5973N mass selective detector (MSD). The experimental array is shown in figure 1.

**Figure 1: Experimental Set-up.**

Cell 1 reactor (figure 1) presents a discontinuity over one of their electrodes and only one dielectric on the top electrode is used, which conserves its flat surface. This first reactor volume is 16 cm × 7 cm endowed with a 0.2 cm gas gap. The reaction volume is approximately 22.4 cm³. The modified electrode exhibits triangular tips on the all its surface and a right angle between the discontinuities (Figure 1, Cell 1). It has been verified by other authors [5] that an electrode with a sharp form improves the generation of free radicals due to the formation of a not-homogeneous electric field above its surface, a field which is more intense on the tip vicinity. Furthermore, the location of the dielectric discharge is sharply defined, that is to say, the associated streamers will converge toward a common point. The second reactor used (Cell 2 in figure 1) is a classic DBDC with double dielectric. Both cell geometries are rectangular and present practically the same dimensions. The power was provided by a high voltage transformer which is fed by half-bridge high frequency inverter [6]. Electric potentials of 10 to 20 kV (p. to. p.) were applied and current levels beyond of 100 mA for the first reactor were obtained. The repetition frequency was maintained constant at 1.75 kHz.

### 3. Results

The small numbers 1 to 6 in brackets seen in the next figures, identify same particular conditions of the rectified direct voltage that is transmitted to the inverter and then to the high voltage transformer. 1 has been used for 90V, 2 for 110V, 3 (120V), 4 (130V), 5 (150V) and 6 (170V).

The evolution of an 80 µmol NO/mol mixture removal in the first reactor is shown in figure 2(a). Figure 2(b) describes the removal in the second reactor with the same concentration and working parameters. The nitrogen oxide concentration was registered each 5s. In figure 2(a, b) an increment of power for reactor 1 with respect to reactor 2 can be observed under the same potential conditions imposed by the power supply. For example, with only a 90 V input (case 1) reactor 1 (figure 2(a)) almost recovers the same 1.63 W power of reactor 2 (figure 2(b)) which requires of 130 V (case 4). This corresponds to 6 kV and 9 kV respectively, with an energy density of 97.8 J/L (0.098 J/cm³) in both cases. It follows that reactor 1 demands a smaller potential in order to produce an energy density equal or greater than that of reactor 2 as a consequence of the use of a single dielectric as well as the tips on the electrode.

Figures 2(a, b) suggest that, if the energy density is increased, so is the removal efficiency, the last one being only slightly higher in reactor 1. One can also observe that, at least at this concentration, the energy density induces an inverse effect on both reactors, i.e., that NO was regenerated. This can be...
explained by the fact that, at high energy density values, nitrogen atoms can be excited and later on produce NO, as shown in the reaction [7]:

\[
N(^{2}D) + N_{2}O \rightarrow NO + N_{2} \quad k = 3.0 \times 10^{-12} \text{ cm}^{3}/\text{s} \text{ at } 300 \text{ K}
\] (1)

As it will be seen later, the appreciable production of NO$_2$ was initially not registered due to the prevailing poor oxygen content, leading us to think on the one hand that, under the working conditions, N$_2$O production took place mainly by means of another mechanism (reaction 2 [7]) different to the decomposition of NO$_2$ (\(N + NO_{2} \rightarrow N_{2}O + O\)) namely

\[
N(^{2}D) + NO \rightarrow N_{2}O \quad k = 6.0 \times 10^{-11} \text{ cm}^{3}/\text{s} \text{ at } 300 \text{ K}
\] (2)

On the other hand, the local heating of the gas can contribute to the regeneration of NO [8], as mentioned in [4] and also observed in our results:

\[
N + O_{2} \rightarrow NO + O \quad k = 4.4 \times 10^{-12} \exp(-3220/T) \text{ cm}^{3}/\text{s}
\] (3)

In the case of a 60 µmol NO/mol mixture, the effect of increasing the energy density and with it the removal intensity is more notorious in the first reactor. Finally, for the 30 µmol NO/mol concentration, reactor 1 presents a higher efficiency for a short time, as seen in figures 3(a) and (b).

The removal efficiencies were calculated from the concentrations ratio \(\frac{n_f - n_i}{n_i}\), where \(n_f\) and \(n_i\) are the final and initial concentration of nitrogen oxide, respectively. The final concentration was taken 50 seconds after the plasma ignition when, for the case of a 30µmol NO/mol mixture in reactor 1, the concentration stabilizes completely. The concentrations are collected and the previous relationship is applied. Then, the 50 second procedure was carried out for reactor 2. The removal efficiencies reached around 98% in reactor 1 and 87% in reactor 2. As follows from figure 3(b), it is possible that similar removal efficiencies are obtained in the reactor 2 albeit with longer times of treatment.

The ion spectra of the 32 µmol NO/mol gas mixture is observed in figure 4(a). One can notice that NO$_2$ is not present both before and after the treatment, while an increment of the abundance of the N$_2$O molecule during the treatment in reactor 1 is exhibited by figure 4(b).

4. Conclusion

All in all, it is clear that reactor 1 has attained an equal or greater energy than reactor 2, under the same described conditions. Additionally, reactor 1 presented a higher efficiency and, during shorter periods of time, also lowers concentrations. In the case of a 30 µmol NO/mol mixture, the observed
efficiency removal reached around 98% for reactor 1 and 87% for reactor 2, after 50s of treatment. At higher 80 μmol NO/mol concentrations, both reactors presented practically the same behavior.

![Graph](image1)

Figure 3: 30 μmol NO/mol removal evolution in Cell 1 reactor (a) and Cell 2 reactor (b).

![Graph](image2)

Figure 4: Ion gas chromatography for NO, N₂O and NO₂ in reactor 1 (a) and reactor 2 (b).

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