

# NOX DEGRADATION ENHANCEMENT IN A DIELECTRIC BARRIER DISCHARGE CELL WITH A SERRATED ELECTRODE

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En este estudio, la superficie del reactor de remoción de Nox, fue modificada con el fin de disminuir el voltaje de la operación, para reducir su nivel de potencial eléctrico y, al mismo tiempo, aumentar su capacidad de remoción, mediante la generación de un plasma frío usando un campo eléctrico no homogéneo en la superficie del electrodo. Este campo eléctrico se consigue por medio de un electrodo de perfil dentado (Reactor 1). Se usaron concentraciones de 30-60 ppm de NOx con 1 L/min de flujo. Los gases de escape se caracterizaron y se analizaron por cromatografía de gases y espectometría de masas. También se llevaron a cabo experimentos adicionales en un reactor con el mismo volumen de reacción pero con dos electrodos convencionales planos (Reactor 2), con el fin de comparar los resultados. La eficiencia de remoción de NOx, tanto con los dos electrodos planos como en la celda DBD aserrada, se acercó al 98 % con la energía más baja en éste último. En ambos casos las medidas experimentales de remoción de NOx se llevaron a cabo en un reactor de volumen 22,4 cm<sup>3</sup>.

Palabras clave: Reactor para el tratamiento de remoción de NOx.

In this study, the electrode surface of NOx removal treatment reactor was modified in order to lower the operation voltage, 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 fields was achieved by means of a serrated electrode profile (Reactor 1). Concentrations of 30-60 ppm of NOx in nitrogen was used with 1 L/min flow. The exhaust gases were analyzed as well as characterised by gas chromatography and mass spectrometry. Additional experiments were also carried out in a reactor of the same reaction volume but where two conventional flat electrodes (Reactor 2), in order to compare the results. The NOx removal efficiency as in the two flat electrodes as in the serrated DBD cell were close to 98% with the lower power in this last one both experimentals removal of NOx were carried out in a 22,4 cm<sup>3</sup> volumen reactor.

Key words: NOx removal treatment reactor.

### Introduction

Recent successful applications of non thermal plasmas include, particularly, processes of gas flow decontamination. One of the main atractives of these techniques is that the gas temperatura doesn't increase substantially/1/ and dangerous secondary compounds like subproducts 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 was treated in a newly

designed reactor where the flat surface of the electrode was modified changing its configuration to serrated in order to produce a controlled dielectric discharge. The removal efficiency obtained was compared with those obtained from a traditional reactor of two flat electrodes with dielectric cover

### **Experimental Set-Up**

A gas mixture of nitrogen oxide balanced with nitrogen gas was selected, with concentrations around 30 and 60 ppm.

The total mixture flow was fixed at 1,0 L/m and the concentration during all the experiments was measured with a Sensonic 2000 gas analyzer.

The electric charge and the voltage in the cell were performed by a tektronic osciloscope TDS 2014 and a high voltage P601A probe for tektronik respectively while the current in the discharge was gauged using a 0,1-1,0 W 1,0V/A transformer from pearson electronics.

The experimental power in the cell during the discharge was calculated using the the lissajous figure (5) The experimental array is shown in figure 1.



Fig. 1

DBD cell with a serrated electrode (figure 2) presents a discontinuity and exhibits triangular tips over one of its metalic surface electrode and a right angle between the discontinuities The other metallic electrode conserves its flat surface and its cover with a dielectric. DBD cell with two flat electrodes conserves its original configuration Both reactors volume is approximately 22,4 cm<sup>3</sup> ( $16 \times 7 \text{ cm}^2 \times 0,2 \text{ cm}$  gas gap)

It has been verified by other authors (6) that electric energy consumed in the discharge increase linearly with the area The electrodes used ain the the serrated electrode DBD cell improve the generation of free radical due to the formation of a more dense non homogeneous electric field above its surface, a fiel wich is more dense in the tip vecinity, it's mean a larger energy by area unit.



Fig. 2

Furthermore, the location of the dielectric discharge is sharply defined, that is to say, the associated streamers will converge toward a common point. It is expected that the use of this type of electrode makes the discharge onset voltage smalller.

The power was provided by a high voltage transformer which is fed by half-bridge high frequency inverter/7/ Electric potentials of 1,3 to 1,6 for the serrated DBD cell and 3,25 to 4,5 kV (p. to. p.) in the flat electrode DBD cell were applied and a repetition frequency was maintained constant at 1,75 kHz for all the experimental evaluation.

The figure 3 show the serrated electrode Metal-Glass Metal (M-GM) DBD cell degradation evolution for 30 and 60 NO ppm concentration and the figure 4 shows its corresponding Q-V Lissajous curves for 60 ppm 150V.

The figure 5 show the same experimental case carried out in a flat electrodes Metal

Senated electrode 16x7x0,2 cm DBD cell



Fig. 3

Glass -Glass Metal (MG-GM) degradation DBD cell and the figure 6 shows the electrical Q-V Lissajous curves characteristics for 60 ppm 150V The oxide nitrogen concentration was registered each 6 s for both experimental cases





#### 3. Results

Tables 1 and 2 shows the specific energy determination for 30 and 60 ppm NO, 120 and 150 voltage as particular conditions of the rectified direct voltage that was transmitted to the inverter and high voltage transformer. As we can see the serrated DBD cell needs a lower specific energy than the flat surface DBD cell to reach similar degradation efficiencies, both cells under the same gas and electric conditions.



Fig. 5

For example a 60 ppm NO, 120 V input, the serrated DBD cell demands only 3,50E-01 J/L (Joule/liter) and the flat electrode surface DBD cell, 4,56E-01 J/L, reaching efficiencies of 95,04 % and 89,40 % respectively.





It follows that reactor 1 demands a smaller specific energy density in order to produce an equal or similar degradation efficiency than reactor 2 as a consequence of the use of a single dielectric (M-GM DBD cell) as well as the tips on its electrode.

The removal efficiencies were calculated from the concentrations ratio.

$$\eta = \frac{[NO_{r} - NO_{r}]}{NO_{r}} x100$$

where:

are the initial and final concentration of nitrogen oxide, respectively. In this work, we ilustrate the final concentration for 5, 15 and 30 lecture corresponding to 0,5, 1,5 and 3 minutes after the plasma ignition was started.

The degradation efficiency was calculated since all the experimental procedure were carried out for both reactors and high removal efficiencies were reached.

Table 1 Serrated electrode 17x6x0,2 cm NOx's degradation DBD cell

NO	30 ppm		60 ppm					
V	120V	150V	120V	150V	volts			
Large	4	5,65	8	9,5	cm			
High	0,7	0,6	0,75	0,75	cm			
Area	2,8	3,39	6	7,125	cm <sup>2</sup>			
Vx	2,00E+03	2,00E+03	2,00E+03	2,00E+03	volts			
Vy	5,00E-01	5,00E-01	5,00E-01	5,00E-01	volts			
Cdie	2,00E-06	2,00E-06	2,00E-06	2,00E-06	farads			
Energy	5,60E-03	6,78E-03	1,20E-02	1,43E-02	joules			
Specific E	1,63E-01	1,98E-01	3,50E-01	4,16E-01	J/L			
% efic degradacion								
Lect 1	32,5	30,5	60,5	61,5				
Lec 5	5	10	18,5	22				
Eficiencia	84,6	67,2	69,4	64,2				
Lect 15	2	0	3	2				
Eficiencia	93,85	100,00	95,04	96,75				
Lect 30	1,5	0	2,5	1,5				
Eficiencia	95,4	100,00	95,8	97,5				

Figure 5 show similar removal efficiencies obtained in the reactor 2 albeit with longer times and great treatment power for all these cases.

Figures 3 and 5 show that in all cases, experimental NO degradation concentration stabilizes more or less after 0,7 minutes (42-45 seconds) and the degradation decrease slowly after de first minute until reach the lower value degradation although NOx regeneration can be possible because electrical field enhancement allows reactions that increase

#### Table 2 Flat electrode 16x7x0.2 NOx's degradation DBD cell

NO	30 ppm		60 ppm						
V	120V	150V	120V	1507	volts				
Largee	2,5	2,3	2,5	2,5	cm				
high	0,7	0,825	1,25	1,275	cm				
Area	1,75	1,8975	3,125	3,1875	cm2				
٧x	5,00E+03	5,00E+03	5,00E+03	5,00E+03	volts				
Vy	2,00E-01	5,00E-01	5,00E-01	5,00E-01	volts				
Cdie	2,00E-06	2,00E-06	2,00E-06	2,00E-06	farads				
Energy	3,50E-03	9,49E-03	1,56E-02	1,59E-02	joules				
Specific E	1,02E-01	2,77E-01	4,56E-01	4,65E-01	jЛ				
% efic degradacion									
Lect 1	30,7	28	61,3	61,5					
Lec 5	16	18	27,3	30,5					
Efic	47,8	35,7	55,4	50,4					
Lect 15	2	0	6,5	1,3					
Eficiencia	93,49	100,00	89,40	97,89					
Lec 30	2	0	6,5	0,8					
Efice	97,8	100	89,4	98,7					

#### NO production /8/

The degradation was completely for the case of a 30 ppm NO mixture and 150 V reaching an efficiency in both reactors, employing a lower power in the serraated DBD cell . In the case of a 60 ppm mixture, and 150 V the efficiency removal reached was 97.5 and 98,7 % respectively for reactor 1 and 2.

## Conclusion

It's clear that reactor 1 in this experimental had attain an equal or high efficiency with a low specific energy than reactor 2 during shorter periods of time, under the same described conditions for example for the first 5 seconds the NO concentration in the serrated DBD cell was 18.5, 22 ppm with 120 y 150 V while in the flat electrodes cell it was 27,3 y 30,5 ppm (It can be read like lect 5 in tables 1 and 2), in fact because the energy was concentred in a minor area and because each tip treatment time in reactor 1 was very short in such a way that at the end of the reactor 1 the treatment time is shorter than reactor 2. For each discharge the energy by area is greater in the serrated cell so it's posible to build smaller reactors with tips in order to work with a lower voltages and specific energy and to obtain the same Nox's degradation high efficiencies like in the flat electrodes conventional DBD cell

## References

- Anderson G, Bechotold L, Greene A, Heck H, Kang M, Tennant R, Vargas R and Wantuck P Report to LANL Advanced Waste Treatment Program (FY92 activities under TTP AL121126). 1992
- 2 Kogelschatz U Dielectric Barrier Discharge: Their History. Discharge Physics and Industrial Applications. Plasma Chemistry and Plasma processing Vol 23 No 1-46 (2003)
- 3 Kim H H, Prieto G, Takashima K, Katsura S and Mizuno A Performance evaluation of discharge plasma process for gaseous pollutant removal Department of ecological engineering Toyoohashi University of technology Tempoku-Cho Japan, J. Electrostatics 55 25 – 41 (2001)
- Penetrante B M, Hsiao M C, Merritt B T, Vogtlin G E and Wallman P H Comparision of electrical Discharge Techniques for Nonthermal Plasma Processing of NO in N2 IEEE Trans.Plasma Sci. 23 679 – 687 (1995)
- 5 I Nagao. M Nishida, K Yukimura S Kambara and T Maruyama. Nox Removal using nitrogen gas activates by dielectric barrier discharge at atmpspheric pressure. Department of electrical engineer Dashisha University 1-3 Tatara Miyakodani kyotanabe, kyoto japan, Vacuum 65 481-487 (2002)
- 6 Koichi takaka, Muaffaq A Jani and Tamiya Fujiwara Renoval of nitric acid in flue gases by multipoint to

plane dielectric barrier dischar ge. IEEE transactions on plasma science vol 27 N° 4 (1999)

- 7 Godoy-Cabrera O G, López-Callejas R, Mercado-Cabrera A, Barocio S R, Valencia R, Muñoz-Castro A, Peña-Eguiluz R and A de la Piedad-Beneitez m A High Frecuency (100kHz) operated ilent discharge cell Plasma Sources Sci. Technol. 15 360 367 (2006)
- 8 A mercado ,Godoy C, Valencia A, Lopez C, Barocio S, Peña E, Muñoz C, De la Piedad B, Del Villar E Nox's Treatment by Dielectric Barrier Discharge. XI Latin American Workshop on plasma Physics AIP Conference Proceedings vol 875, pp 236-239(2006)