Three-electrode plasma reactor for the removal of toxic gases

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Abstract. Electrical and spectroscopic measurement for the characterization of a novel three-electrode plasma reactor for the treatment of toxic gases is presented. The three-electrode discharge consists in a dielectric barrier discharge (DBD) combined with a corona discharge (CD). The DBD is generated by applying an alternating high voltage signal between two circular aluminium plate electrodes attached to opposite sides of a disk made of dielectric material. The CD is generated applying a continuous negative high voltage to an external cylindrical mesh electrode, coaxial with the DBD electrode system. The gap between the edge of the DBD system and the mesh electrode is approximately 20 mm wide. Up to five DBD electrode systems can be connected in parallel inside the reactor, axially separated from each other by 30 mm. The electrical characterization consisted in the measurement of the current between the DBD system and the external mesh, and the voltages of the electrodes. In order to understand the dynamics of the streamers, a theoretical determination of the laplacian electric field generated by the biased electrodes was done. Optical emission spectroscopy was performed in the range of wavelengths 280-480 nm, containing the typical spectral bands 2nd positive and 1st negative systems of molecular nitrogen.

1. Introduction
The emission of gases containing pollutants by industries, automobiles, and others are a big environmental problem in today’s world. To remedy this problem, many techniques have been studied by several scientific teams. Techniques employing plasmas at atmospheric pressure have been very successful in removing toxic molecules present in polluted gases. Some of them are DBD and CD [1, 2]. Plasmas formed in these discharges are non-thermal plasmas in which energetic electrons collide with gas molecules and produce a rich variety of chemical products that undergo reactions capable of transforming harmful substances into non-hazardous products.

In a previous work [3], the development of a coaxial three-electrode plasma curtain discharge was presented. The discharge was based on the combination of a DBD with a CD in a three-electrode system with cylindrical geometry; basically, it resulted from the ‘stretching’ of a pure DBD discharge by the action of a CD discharge generated between the active electrode of the DBD and a remote third electrode. This discharge configuration, with double DBD electrode systems connected in parallel,
produces almost double average current than that corresponding to the single one (for the same voltage values), indicating that in the stacked arrangement, both discharge systems develop independently.

In this work, we present a three-electrode plasma reactor for the removal of toxic gases. Electrical characterization was carried out, with air flowing in the reactor, by measuring the interelectrode current. In order to understand the dynamics of the streamers a theoretical determination of the laplacian electric field generated by the biased electrodes was done. Also, a plasma emission spectrum of air was obtained to observe the different species produced in the plasma.

2. Experimental

A schematic of the experimental setup is shown in Figure 1. The three-electrode reactor arrangement consists in two disks of adhesive aluminium tape of 50 μm thickness and 19 (electrode 1) and 17 (electrode 2) mm of radius, flush mounted at both sides of a poly-methyl methacrylate dielectric disk of 40 mm diameter and 2 mm width. One of the disks is connected to an ac power supply and the other to ground by wires placed inside two insulating tubes (10 mm in diameter) which pass along the electrode system. A third electrode (electrode 3), consisting in a steel mesh connected to a dc power supply is attached to the inner wall of a poly-methyl methacrylate dielectric cylindrical tube with 80 mm inner diameter, 240 mm length and with a wall thickness of 5 mm. This tube surrounds the central electrode arrangement and the distance between the edges of the central electrodes and the third electrode is 20 mm. Two plates seal the ends of the tube. Up to five DBD electrode systems can be connected in parallel inside the reactor. The gas, under pressure, goes through the reactor. The gas inlet is placed at one plate and the gas outlet in the other plate.

For our circuit, the optimum excitation ac frequency was 5.3 kHz. The ac voltage amplitude (Vac) was 6 kV and Vdc was -12 kV. The Vac was measured using a high voltage probe (1000 X / 3.0 pF/100MΩ). Current measurements were inferred from the voltage drop through a 50 Ω resistance. These electrical signals were registered with a four-channel digitizing oscilloscope with a bandwidth of 60 MHz and 1 Gs/s of sampling rate. The air flow was measured using a stainless steel float flow meter in the range 0 -17 l/min. Spectroscopy measurements were performed with a monochromator (280-480 nm) attached to a photomultiplier (Hamamatsu 1P28). Figure 2 shows a photograph of the reactor.

![Figure 1. Schematic of three-electrode reactor. 1 and 2 are the DBD electrodes and 3 is the external electrode.](image-url)
3. Results

These In Figure 3 Typical signals of the voltage $V_{ac}$ (a) and the interelectrode current $I$ (b) for five DBD electrode systems without air flow, are presented. Pulses superposed on the capacitive current indicate the presence of streamers crossing the electrode gap. In our configuration, the ignition of the streamers starts at $V_{ac} = 2$ kV, and the extinction of the streamer occurs when the peak value $V_{ac} = 6$ kV is reached. To derive some meaningful information on the discharge characteristics the interelectrode current is presented as an average value ($I_{m}$) by setting the oscilloscope in the average acquisition mode, an average waveform of the current signals over 128 samples were acquired. Then, to ignore the reactive component of the current these statistical-averaged signals were time averaged over one period of the AC voltage to finally obtain the average current values. According to this described procedure the uncertainty in the average current values could be estimated to be around 10%, due to statistical fluctuations.

![Figure 2. Photograph of the plasma reactor.](image)

![Figure 3. Typical signals of the voltage $V_{ac}$ (a) and interelectrode current (b).](image)
In Figure 4, the average interelectrode current, as a function of air flow values (with different number of DBD electrode systems connected in parallel as a parameter) is presented.

![Figure 4](image.png)

**Figure 4.** Current as a function of the air flow for different number of DBD electrode systems for $V_{ac} = 12$ kV pp, $V_{dc} = -12$ kV.

In Figure 5 an emission spectrum of the discharge in air is presented. A typical emission lines of nitrogen was found in the wavelength range $280 – 480$ nm. The relevant lines correspond to the second positive system of the nitrogen molecule.

![Figure 5](image.png)

**Figure 5.** Typical emission spectra,

4. Electrical model
To understand the dynamics of the streamers, the electric field $E$ was evaluated using the analytical expression of the potential for a surface charge density inside a cylinder at a fixed potential [4].

$$
\phi(r, z) = \frac{1}{\pi \varepsilon_0} \left[ \int_0^a \sigma(r') r' I_0(kr') dr' \right] \cos[k(z-z_d)] \left[ K_0(kr) - I_0(kr) \frac{K_0(ka)}{I_0(ka)} \right] dk
$$

(1)

where $a$ is the radius of the external electrode, $a_d$ the radius of the surface charge distribution, located at the axial position $z_d$, $\varepsilon_0$ is the permittivity of the vacuum, $K_0$ and $I_0$ the modified Bessel functions of order 0. Expression (1) is valid for any axial coordinate $z$, and for radial coordinates $a_d \leq r \leq a$. For a single metallic disk of radius $a_d$ lying on the surface of a dielectric of relative permittivity $\varepsilon_r$, the total surface distribution of electric charge, including the charge on the disk plus the polarization charges on the dielectric surface, is given by [5]

$$
\sigma(r) = \frac{q_d}{2\pi \varepsilon_r a_d^2 \sqrt{1 - r^2 / a_d^2}}
$$

(2)

where $q_d$ is the electric charge in the disk only. Use of the expression (2) in (1) results in the analytical expression for the potential due to a disk on a dielectric surface, in which the integral is to be done numerically. The total potential was obtained as the superposition of the above expression for each DBD electrode. This expression, when evaluated at each disk position, allows to relate the disk charges with the disk voltages (which are the actual inputs), and the electric field is finally obtained by numerical spatial derivation.

In Figures 5 and 6 the potential and electric field as a function of $r$ for different Vac and external voltage $V_{dc} = -12$ kV is presented. According to Raizer [6], to ensure the streamer propagation, the electric field value necessary for the streamer propagation across the air at atmospheric pressure is 4 kV/cm approximately. In Figure 6 can be observed that this value is obtained for Vac values of the figure 3 at which the streamers are generated.

**Figure 5.** Potential as a function of radius $r$ for different Vac and $V_{dc} = -12$ kV.
Figure 6. Electric field as a function of radius $r$ for different $V_{ac}$ and $V_{dc} = -12$ kV.

5. Final Remarks
We have presented a novel three-electrode plasma reactor for the removal of toxic gases. The discharge is stable in a wide range of air flows and for different number of DBD electrodes systems connected in parallel. The main advantage of the geometry of the reactor is that a large plasma volume can be established and presents a natural boundary for the gas flow.

For the applied voltages it was found that the calculated values of the electric field are sufficient to sustain the streamer propagation along the interelectrode gap only during a part of the positive cycle of the $V_{ac}$ voltage in accordance with the experimental observation.

The obtained spectra show the presence of excited molecular $N_2$, indicating the presence of energetic electrons generated by the streamers crossing the interelectrode gap.

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