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Study of Dielectric Barrier Discharges in Nitrogen and Afterglows at Atmospheric Pressure

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Abstract. This article is devoted to the study of Dielectric Barrier Discharges (DBD) and afterglows obtained at atmospheric pressure in nitrogen. Electrical and optical analysis of the DBD reactor are carried out in order to characterize specific activated gaseous species, which can travel with low energy dispersion for long distances in tubes. A hypothesis for the explanation of the presence of long-lived species in such afterglows is finally explained as related to a solitary wave effect.

1. Introduction
Dielectric Barrier Discharges (DBD) are currently used in order to produce non-thermal plasmas at atmospheric pressure. Introduced for the first time by Siemens [1], DBDs consist in depositing on, or between HV and grounded electrodes, one or two dielectrics in order to avoid arc transitions. Depending on the gas and on the HV power supply used, such discharges are usually multi-filamentary but can exhibit diffuse characters [2].

In this work is shown a DBD reactor working in nitrogen and supplied by a pulsed-like HV generator. After a brief description of the set-up, experimental diagnostics allow characterizing the excited gas and a particular attention is given to the afterglows obtained by this way. At last a particular explanation of the formation of luminous afterglows in tubes at atmospheric pressure is given supposing a solitary wave effect.

2. Experimental set-up
Figure 1 shows a schematic representation of the experimental set-up. It can be seen that it is possible to connect an adaptor at the exit of the DBD reactor in order to channel the gas and obtain afterglows at atmospheric pressure.

The DBD reactor has been conceived in a coaxial configuration with a gap of 1mm and a discharge area of 1cm in length. The inner electrode is supplied by a pulsed-like HV generator and the outer is grounded. An alumina coating has been applied on the HV electrode in order to cover it by a dielectric with constant permittivity (ɛ = 9). Rounded profiles have been machined on the grounded electrode in order to avoid point/edge effects (local enhancement of the Electric Field) between the extremities of the inner electrode where no dielectric coating is applied and the grounded electrode as it is shown in figure 1 (see the sketch of the DBD reactor).

The HV generator is a bipolar pulsed-like supply which delivers high voltage pulses with residual oscillations as it can be seen on figure 2a. Nevertheless, this HV power supply allows adjusting numerous parameters as:

- the frequency in the [8–180 kHz] range,
- the HV amplitude in the [0–8 kV] range,
- the rise times in the [0.2–0.6 kV/ns] range,
- the pulse duration in the [1–5 μs] range.

Figure 1. Experimental set-up and sketch of the DBD reactor.

The applied voltage $v_m(t)$ is measured by a 1/1000 Tektronik P6015A voltage probe while the total current $i_m(t)$ flowing in the circuit is measured by a rogowski-type current probe (Tektronix CT-2). The electrical signals are visualized by means of a Tektronix 3054B oscilloscope (500 MHz, 5 Gs/s). By using an electrical equivalent of the system (figure 2b) it can be deduced from the measurements of the delivered voltage $v_m(t)$ and of the total current $i_m(t)$ the gap voltage $v_g(t)$, the discharge current $i_d(t)$, and the plasma power $p(t)$.

Figure 2a. Electrical voltage, current, and power waveforms for the positive pulse of the bipolar HV pulsed-like generator.

Figure 2b. Electrical equivalent of the DBD reactor ($C_d$: dielectric capacity, $C_g$: gas capacity, $Z_g$: dissipative component in the gas).

This way, the mean plasma power can be deduced during one period of the electrical signals and related to the discharge surface: the mean plasma power density can thus be adjusted in the [1–50 W/cm²] range.
The gas used is nitrogen and is delivered by a cryogenic vessel (purity 99.995 %) in the axial
direction of the electrodes. Due to the configuration of the experimental device, the gas is evacuated in
the working room so that the gas is composed of oxygen impurities. The gas flow is controlled by a
gas flowmeter at the inlet of the reactor and can be varied from 1 to 100 slm.

Optical analysis of the discharge and of the afterglow have been conducted with a CCD equipped
JY-1000M spectrometer with a 1200 gr/mm grating blazed at 500 nm. In each case, a fiber optic is
used and the light is focalized by means of an optical lens.

3. Optical Emission Spectroscopy
The analysis of the inside of the discharge by emission spectroscopy indicates mainly the presence of
the Second Positive System (SPS) of the molecular nitrogen, as shown in figure 3.

![Figure 3. OES diagnostic inside the nitrogen DBD.](image)

The N₂ SPS emissions are due to the following reaction:
\[
N_2(C) \rightarrow N_2(B) + h\nu
\]
and its production is characteristic of electronic collisions in the gap with molecular nitrogen. This
known result [3–5] is confirmed with the following figure 4 where the correlation can be established
between the main emission of the SPS system at 337.1 nm and the mean plasma power. The higher the
applied voltage the highest the mean plasma power and, consequently, the highest the electronic
collisions in the gas which finally produce higher concentrations of the molecular state N₂(C).

Moreover on figure 3 it can be seen the presence of the First Negative System of the molecular
nitrogen which is characteristic of ionic charges in the gap [3–5] and confirm the interpretation of the
N₂ SPS.

Finally in the UV part of the spectrum, figure 3 indicates the presence of NOγ emissions which
may be due to the oxygen impurities naturally contained in our atmospheric DBD reactor but which
are characteristic of the presence of the metastable excited stated N₂(A) of molecular nitrogen by the
following mechanisms [3–5]:
\[
N_2(A) + NO \rightarrow NO(A) + N_2
\]
\[
NO(A) \rightarrow NO(X) + h\nu.
\]

The addition of a metallic adaptor at the exit of the DBD reactor allows to connect a quartz glass
tube of several mm in diameter and to channel the excited gas (figure 1). It is remarkable that by this
way it is possible to obtain luminous afterglows of several meters at atmospheric pressure and in
numerous materials like polymers, glass, quartz and metallic too. Photo 1 exhibits a nitrogen afterglow
obtained in a silicone supple tube of 1 meter in length.
It can be noticed that this afterglow is produced at low temperature and consequently can be applied for useful applications of biological interest like decontamination and sterilization in order to obtain killing effect on parasites, bacteria, fungi, etc [6–10].

The diagnostic of the afterglow by optical emission spectroscopy allows identifying the species contained in such channel tubes. Figure 5 is a spectrum obtained in a quartz tube of 10 mm in diameter and 50 cm in length. It is shown like in figure 3 the N$_2$ SPS which is characteristic of the reaction (1). Nevertheless in afterglows the gas is in remote conditions far from the discharge so that it can be considered that no charges are still present in order to produce another plasma. The mechanism supposed to obtain the N$_2$ SPS is thus the pooling of the metastable N$_2$(A) as described in reaction (4) [11]:

$$N_2(A) + N_2(A) \rightarrow N_2(C) + N_2.$$  \hspace{1cm} (4)

This is confirmed by the fact that the N$_2$ First Negative System is now not observed in such conditions.

In the UV part of the spectrum appears again the characteristic emission bands of the NO-γ system but also of the NO-β system which is an indicator of the presence of nitrogen and oxygen atoms:

$$N + O + N_2 \rightarrow NO(B) + N_2$$  \hspace{1cm} (5)

$$NO(B) \rightarrow NO(X) + h\nu$$  \hspace{1cm} (6)

Specific characteristic bands denoting the presence of impurities like NH and CN components can also be detected. The formation of such molecules can be explained by the presence of water and by carbon contamination in the set-up from the DBD reactor to the quartz tube.
Figure 5. OES diagnostic of the nitrogen afterglow.

But in comparison with figure 3, new bands appear which have been identified to correspond to the emissions of the First Positive System of molecular nitrogen [3–5]:

\[
N + N + N_2 \rightarrow N_2(B,11) + N_2 \quad (7)
\]  
\[
N_2(B,11) \rightarrow N_2(A,7) + h\nu . \quad (8)
\]

FPS emissions allow determining the nitrogen atom concentration in the afterglow as it has previously been achieved by the NO-titration method [12–13]. In our conditions the atomic nitrogen concentration has been estimated to be comprised between \(4 \times 10^{14}\) and \(10^{15}\) cm\(^{-3}\) and it can be deduced the atomic oxygen concentration which is comprised here between \(10^{10}\) to \(10^{11}\) cm\(^{-3}\). In comparison with the work of Pointu et al. [14] using another atmospheric pressure reactor, it seems we obtain similar concentrations. Finally figure 5 indicates the presence of the aurora green band characteristic of the excimer \(N_2O(1S)\) as described in reactions (9) and (10):

\[
O(1S) + 2N_2 \rightarrow N_2O(1S) + N_2 \quad (9)
\]  
\[
N_2O(1S) \rightarrow N_2 + O + h\nu . \quad (10)
\]

This excimer band progressively appears in the quartz tube denoting a transient phenomenon before the establishment of a steady state regime. This phenomenon has been previously described [15] and seems to be due to a progressive treatment of the tube walls allowing the channelling of the afterglow with low energy dispersion and explaining why it is possible to obtain so long afterglows at atmospheric pressure.

By using an analogy with hydrodynamics solitary laws obtained for the description of the blood flow in vessels of human bodies or for the description of the ‘Mascaret’, it is possible to describe the afterglows by a solitary wave effect. (‘Mascaret’ is a natural phenomenon observed in a few rivers and corresponds in the formation of a spectacular wave with constant amplitude moving against the river’s flow). In order to analyse the phenomenon, it has been observed that the transient regime of afterglow formation depends of the nature of the tube, of its dimensions, and of the surface state of the internal walls. X-photoelectron spectroscopy (XPS) on the internal walls of the quartz tube have shown an increase of the O/C ratio by a factor \(\sim 4\). Consequently it may be assumed that the transient phase progressively treats the tube walls in order to form a new gaseous channel called ‘soliton tube’, as shown in figure 6, in which the excited species may travel with very low energy dispersion. By this way surface recombination of the excited state are probably minimized and only the volume recombination mechanisms could participate to the extinction of the luminous afterglow. Nevertheless afterglows exhibit luminous activity after several meters in the tube. Thus in our conditions, volume recombination is not so consequent. The explanation suggested is that the gaseous channel formed in
the tube allows conserves an important quantity of metastables like N$_2$(A) which glide without being destroyed.

![Diagram of soliton tube](image)

**Figure 6.** Formation of the “soliton tube” (M, M*: gaseous species, excited or not, p: gas pressure).

By this way it is possible to have an analytical solitary description of the excited gas flow in the tube, as indicated in equation (11):

$$\frac{\partial A_1}{\partial \eta} + \frac{3}{2} A_1 \frac{\partial A_1}{\partial \chi} - \frac{1}{2} \frac{\partial^3 A_0}{\partial \chi^2} = 0.$$  \hspace{1cm} (11)

In this Korteweg-de-Vries type equation, dimensionless parameters correspond to: $A_1$ the section of the soliton tube, $\eta$ to a temporal dimension and $\chi$ to a spatial dimension. The low non linear term of this equation compensates the low energy dispersion and consequently may permit to form such systems like afterglows at atmospheric pressure.

**9. Conclusion**

In this work DBD and afterglows obtained at atmospheric pressure in nitrogen have been experimentally studied. Specific nitrogen and oxygen excited species have been identified and quantified and the obtained results are in agreement with similar studies [14]. These analysis have been completed with a soliton model in order to describe the afterglows behaviours with low energy dispersion and conservation of the long-lived excited species like the metastable state of nitrogen N$_2$(A).

**References**