Memory effects in Atmospheric Pressure Townsend Discharges in N_2 and air

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This work is focused on the study of the memory effects in Atmospheric Pressure Townsend Discharges in nitrogen (and air) through the description of electrical measurements on a plane-to-plane DBD. The literature suggests that, in nitrogen, the memory effect is mainly based on $N_2(A^3\Sigma_u^+)$. However, it can only explain the discharge behavior in pure nitrogen and only when the time off between two discharges is lower than the $N_2(A^3\Sigma_u^+)$ lifetime. As reported in previous paper, we have shown that in presence of oxidizing gas, the associative ionization of $N(^2P)$ and $O(^3P)$ could explain the increase of seed electrons and thus stability of the homogeneous discharge. However, if the *time off* is larger than the $N_2(A^3\Sigma_u^+)$ lifetime, the creation of seed electrons cannot be related to metastables created during the previous discharge. An hypothesis is that in this case, the seed electrons can come from spontaneous electron desorption, a process that is generally of minor importance at high frequency but which could become predominant at low frequency.

1. Introduction

Dielectric Barrier Discharges (DBD) have tremendous popularity for atmospheric pressure applications including thin-film coating, sterilization, treatment of flue and toxic gases, aerodynamic flow control, and energy-efficient lighting devices. Depending on the gas, electrical parameters, and electrode configuration, these discharges can operate in the classical filamentary mode or in a homogeneous mode. The filamentary mode can be very restrictive for some applications (*e.g.* surface coating). On the contrary, conditions to get a homogeneous DBD can also be restrictive. Homogeneous DBD at atmospheric pressure have been obtained in helium, argon, nitrogen [1]. In nitrogen, the ionization level is too low to allow the formation of a cathode fall. Thus the electrical field is quasi-uniform over the discharge gap, like in low-pressure Townsend discharge, and the obtained discharge is called Atmospheric Pressure Townsend Discharge (APTD) [2]. For a Townsend breakdown to occur, a production source of secondary electrons is necessary when the electric field is low. The aim of this work is to better understand the mechanisms that could be at the origin of the production of seed electrons in pure nitrogen and in air.

2. Experimental set-up

The experimental set-up has already been described in previous publication [3]. The DBD is kept in a closed vessel to perform experiments in a very well controlled atmosphere. The plasma reactor is slightly pumped down to 10^{-3} mBar before each experiment. The DBD is ignited at atmospheric pressure between two parallel electrodes (3×3cm^2) made from metalized paint deposited on 635µm thick alumina plates (115×70mm^2, relative permittivity of 9.6). The gas gap is 1mm. Epoxy resin covers the electrodes and the electrical contacts to ensure electrical insulation of the whole device.

The power supply is a high voltage linear amplifier whose output is connected to the cell discharge.

In order to renew the atmosphere, a 2 slm total gas flow of pure nitrogen or synthetic air (Air Liquide Alphagaz 1, 99.9998 purity) is injected from one side of the discharge (longitudinal gas injection), keeping a constant pressure of 1 bar through a gentle pumping of the vessel.

The discharge is characterized by electrical and light measurements. The voltage applied to the electrodes is measured by means of a high voltage probe (Tektronix P6015A). The discharge current is measured through a shunt resistor in series with the electrodes. The current and the voltage applied to the electrodes are visualized on a numerical oscilloscope (LECROY WaveRunner HRO 66Zi) through coaxial cables. The discharge homogeneity is investigated by means of short exposure time pictures, which are taken with an intensified CCD camera (PI-MAX 3, Princeton Instruments) synchronized with the power supply voltage.

3. Memory effect in DTPA

When the discharge is in the Townsend mode, it is characterized by electrical characteristics (discharge current and voltages) shown on figure 1.

The current I_d consists of a lone pulse per half cycle. Its shape is reproducible from one discharge to another. The gas voltage plateau is characteristic of the Townsend discharge. An important feature is the fact that the discharge current never reaches zero between two discharges. Hence a current jump can be measured when the gas voltage polarity reverses. This current is due to seed electrons generated in between two discharges, when the field is low enough to "trap" them in the gas volume, and thus is a clear signature of the memory effect from one discharge to the other.

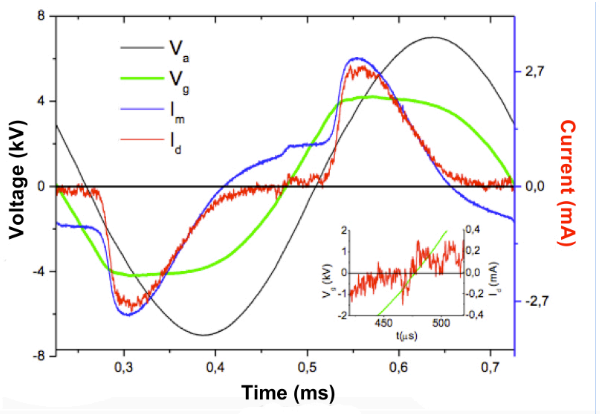

Fig. 1: Voltage and current waveforms of an APTD in N_2. V_a is the applied voltage, V_g the gas voltage, I_m the measured current and I_d the discharge current (f=2 kHz, V=14 kV_{pp})

The importance of this memory effect is also pointed out by the evolution of the first discharges after ignition (Figure 2). The first two discharges are different from the following ones showing that one DBD pulse depends on the previous one and that some discharges are necessary to reach the equilibrium. The first discharge is filamentary, the second one is a mix of a Townsend discharge and micro-discharges and the third one is fully a Townsend one.

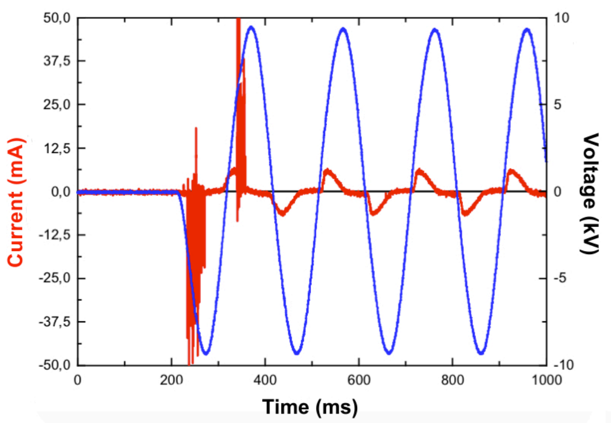

Fig. 2: Voltage and current waveforms of an APTD in N_2 during the first discharges

In nitrogen-based discharges the creation of seed electrons is correlated with the presence of $N_2(A^3\Sigma_u^+)$ metastable molecules created during the previous discharge and persisting between two discharges [4]. These seed electrons could come from secondary electron emission by $N_2(A^3\Sigma_u^+)$ impact on the dielectric surface [2]. As metastables can induce secondary emission, this mechanism can continuously produce electrons between two discharges. This phenomenon would be favor by the fact that before the breakdown, the new cathode is negatively charged, which enhances the γ coefficient as it has been recently shown [5]. However, this mechanism can not explain on its own the discharge behavior observed when (i) a few tenth of ppm of O_2 are injected in the gas, (ii) the discharge is obtained in air (Figure 3), (iii) the frequency is much lower than 1 kHz (Figure 4).

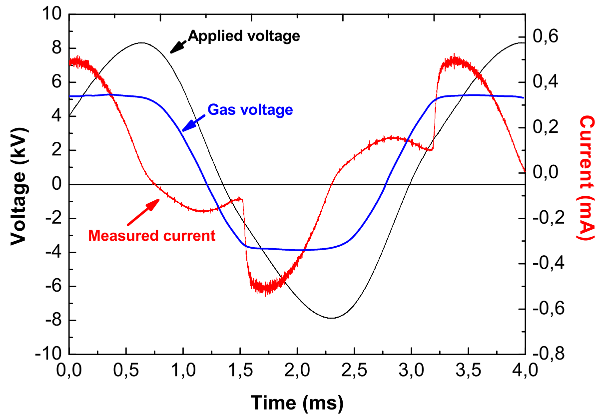

Fig. 3: Voltage and current waveforms of an APTD in air (f=300 Hz, V=17 kV_{pp})

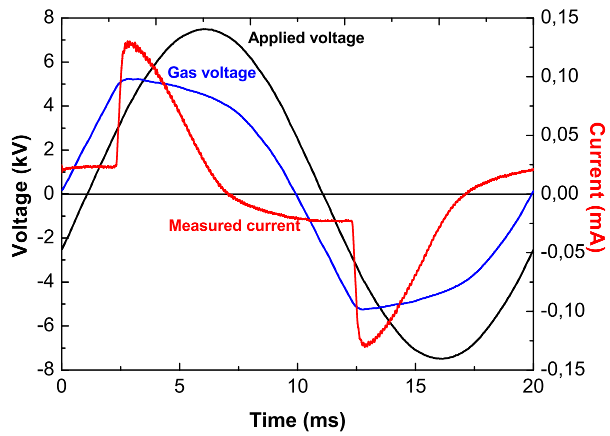

Fig. 4: Voltage and current waveforms of an APTD in N_2 at low frequency (f=50 Hz, V=15 kV_{pp})

As reported in [4], the current jump strongly increases when small amount of O_2 (< 100 ppm) is added to N_2. Then, the number of seed electrons increases whereas the $N_2(A^3\Sigma_u^+)$ density strongly decreases due to quenching effects [4]. In presence of a small amount of oxidizing gas, the associative ionization of $N(^2P)$ and $O(^3P)$ could explain this effect [6]. However, at low frequency, and for much high O_2 amount (up to 20%, see Figure 3) the discharge can still be homogeneous and in Townsend regime. Therefore, in this condition the mechanisms for the creation of seed electrons between two discharges, which is essential to generate a stable homogeneous discharge, can not be based on the $N_2(A^3\Sigma_u^+)$. We can notice that in air, the Townsend discharge is only obtained at low power and thus low frequency (f < 1.5 kHz).

In nitrogen, as we can see from figure 4 the discharge can still be homogeneous and in Townsend regime for frequency of only a few tens of Hertz [7]. When the frequency decreases, the time between two discharges decreases too and can become higher than the $N_2(A^3\Sigma_u^+)$ lifetime. For example, in pure nitrogen, LIF measurements show $N_2(A^3\Sigma_u^+)$ densities as high as 10^{13} /cm^3, with a lifetime around 40µs [8]. Consequently, at low frequency the mechanisms for the creation of seed electrons between two discharges can not be based on the $N_2(A^3\Sigma_u^+)$.

However, even at low frequency, the first discharge is always filamentary which is an evidence of a memory effect persisting between two discharges. Therefore, when this memory effect can not be related to the presence of $N_2(A^3\Sigma_u^+)$, another mechanism generate seed electrons. This mechanism works only at low frequency and thus we studied the influence of the applied voltage slope.

4. Effect of the slope of the power supply voltage

To obtain a Townsend discharge, the ionization has to be slow enough to avoid a large avalanche development. The ionization speed is related to the slope of the power supply voltage (*dV/dt*). As we can see from figure 5, with a slow slope for the rise time of the applied voltage, the discharge is homogeneous whereas the discharge is filamentary on the fall time of the applied voltage due to the high slope. Consequently *dV/dt* is a key parameter to obtain a homogeneous DBD.

To study the influence of the *dV/dt* on the discharge in pure nitrogen, we use a triangular voltage with a variable duty cycle in order to adjust the rise and fall times. The voltage applied to the DBD is a burst of two periods of this signal followed by an adjustable off time (Figure 6).

Then, for different frequencies of the triangular voltage (50, 100, 120, 150, 400 and 1000Hz) and different *off time* (up to 50 ms), we determined the maximal value of the *dV/dt* for the first discharge after the *off time* to obtain a homogeneous discharge (Figure 7).

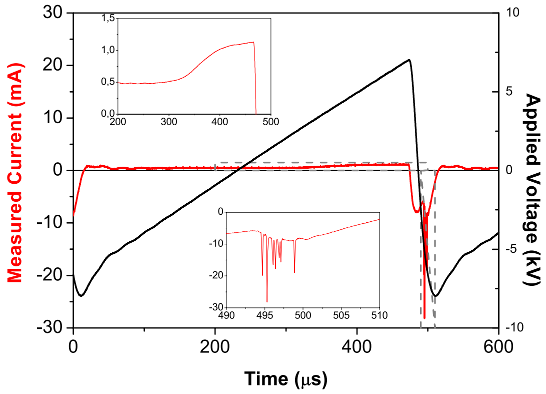

Fig. 5: Influence of the slope of the power supply voltage (*dV/dt*) on the discharge regime (f=2 kHz, V=15 kV_{pp})

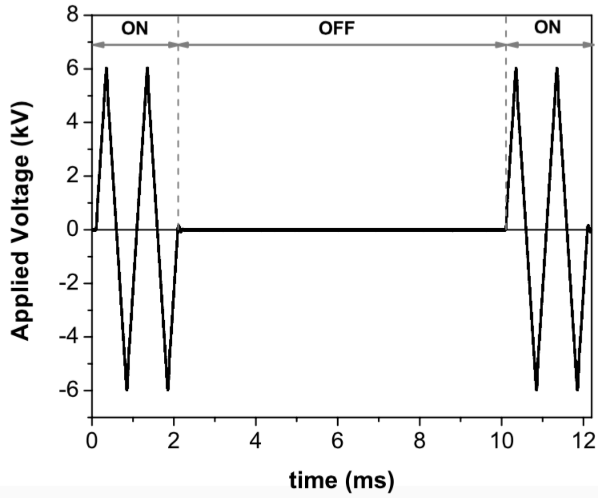

Fig. 6: Burst triangular voltage applied to the cell discharge

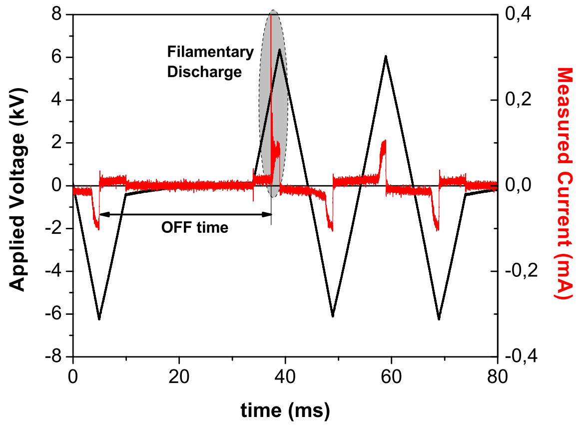

Fig. 7: Voltage and current waveforms using the triangular burst signal (f=50 Hz, V=12 kV_{pp})

As we can see from figure 8, the higher the *off time* between two discharges is, the lower is the maximal value of the voltage slope allowing to obtain a homogeneous discharge. This suggests that a high *dV/dt* during the breakdown requires a strong memory effect and therefore many seed electrons. This is confirmed by the addition of points corresponding to the maximum power (5 W/cm^2, f=8-10 kHz) obtained with a sinusoidal voltage (figure 8). It is interesting to note that all the points are on the same slope.

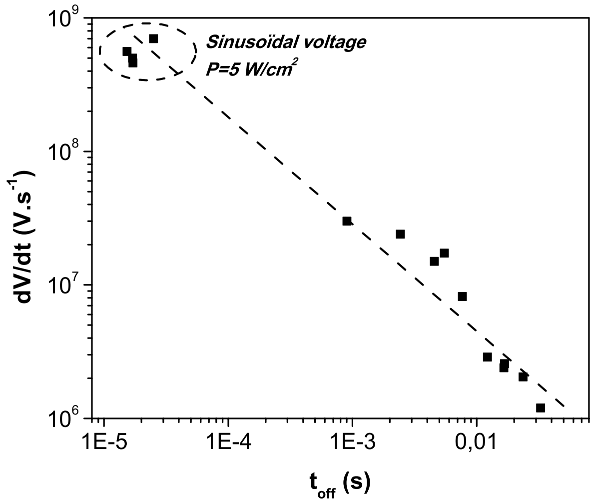

Fig. 8: Maximal value of dV/dt for a homogeneous discharge as a function of the *off time*

5. Discussion

If the *time off* is larger than the $N_2(A^3\Sigma_u^+)$ lifetime, the creation of seed electrons cannot be related to metastables created during the previous discharge. Therefore, there must be another mechanism to produce the seed electrons. As we have seen, the first discharge could be homogeneous with a *off time* up to 30 ms. Thus, the time constant of this mechanism must be of the order of ms.

Then, an hypothesis is that the seed electrons can come from a spontaneous electron desorption [9]. In pure nitrogen for frequency higher than 1 kHz, the electron desorption process is of minor importance [10], however at low frequency and in air it could be the major process. This could explain why a homogeneous discharge in air is only obtained at low frequency.

Based on this hypothesis, the figure 9 presents a schematic diagram of the mechanisms at the origin of the memory effect in nitrogen and nitrogen/oxygen mixtures as a function of the experimental conditions.

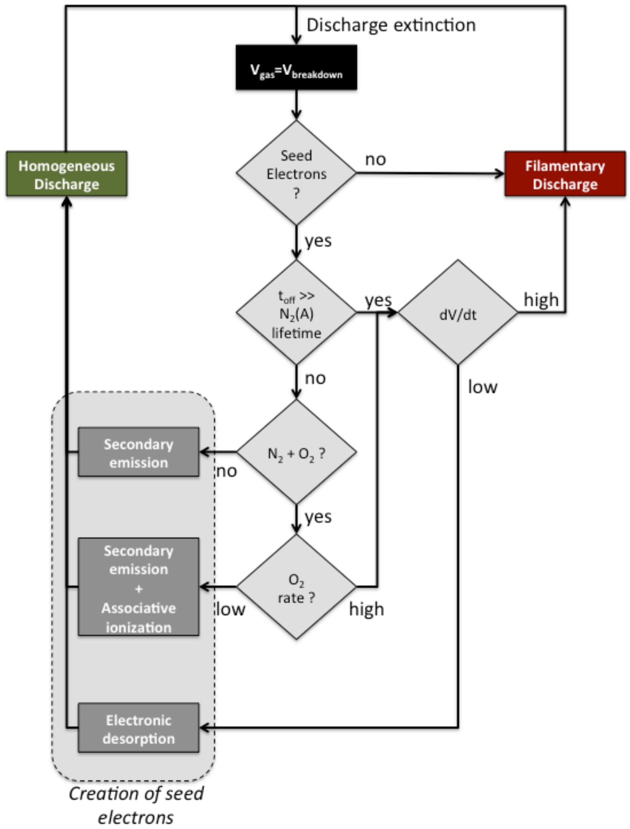

Fig. 9: Schematic diagrams of the mechanisms at the origin of the memory effect in N_2 Townsend discharge at atmospheric pressure

6. References

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