Atmospheric Pressure Townsend Discharges (APTD) in various molecular gases: pre-ionisation mechanisms?

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Abstract: This work is focused on studying the pre-ionisation mechanisms in Dielectric Barrier Discharges (DBD) at atmospheric pressure in molecular gases and mixtures leading to a homogeneous Townsend discharge. The seed electrons required for the formation of a Townsend discharge can originate from the surfaces or the gas bulk. This work aims to summarise and quantify the importance of the predominant sources of seed electrons, considering the different memory effect mechanisms.

Keywords: DBD, Townsend, pre-ionisation, volume, surface, mechanism.

1. General

Dielectric Barrier Discharges (DBDs) can be used for many atmospheric pressure applications, including thin-film coating, sterilisation, treatment of flue and toxic gases, aerodynamic flow control, and energy-efficient lighting devices [1-3]. Depending on the gas, electrical parameters, and electrode configuration, these discharges can operate in the classical filamentary mode or in a homogeneous mode [4-5]. The filamentary mode can be very restrictive for some applications (*e.g.* surface coating). Nevertheless, conditions to get a homogeneous DBD can also be restrictive. Homogeneous DBDs at atmospheric pressure have been obtained in helium, argon, and nitrogen [5]. In nitrogen, the ionisation 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 discharges, and the obtained discharge is called Atmospheric Pressure Townsend Discharge (APTD) [5]. For a Townsend breakdown to occur, a production source of secondary electrons is necessary to sustain the APTD when the electric field is low. This work aims to synthesise the mechanisms that could be at the origin of the production of seed electrons in various molecular gases and to understand how to favorise the obtention of a Townsend breakdown.

2. Experimental setup

The experimental setup has already been described in previous publications, e.g. in [6]. The DBD is kept in a closed vessel to perform experiments in a well-controlled atmosphere. The plasma reactor is pumped down to 10^{-3} mbar before any experiment and then is filled to atmospheric pressure using mixtures of gases (99.999% purity) purchased from Air Liquide. The discharge is ignited between two alumina plates separated by a 1 mm gas gap, and the discharge area is 3x3cm^2. To renew the atmosphere, a gas flow (1 slm) 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 sinusoidal high-voltage power supply consists of a low-frequency generator providing the reference waveform, amplified by a linear amplifier whose output is applied to the primary winding of a transformer in series with a 4 Ω resistor. The electrodes are connected to the secondary winding of the transformer. The DBDs are characterised by electrical measurements. The high voltage applied to the electrodes is measured using a high-voltage probe. The discharge current is measured through a 200 Ω resistor in series with the electrodes. The current and the voltage applied to the electrodes are visualised on a digital oscilloscope. The discharge homogeneity is investigated using short exposure time pictures, which are taken with an intensified CCD (iCCD) camera synchronised with the applied high-voltage waveform.

3. Townsend discharge at atmospheric pressure

The physics of diffuse discharges obtained in atmospheric pressure DBD in nitrogen is well known even in mixtures containing oxygen species [5,7-8].

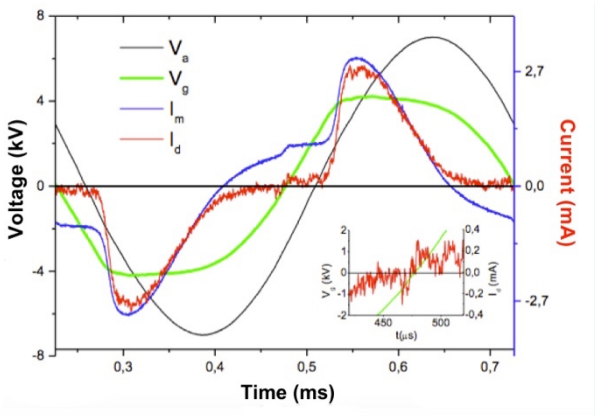

Fig. 1. Voltage and current waveforms of an APTD in N_2. V_a is the applied high-voltage, V_g the gas gap voltage, I_m the measured current and I_d the discharge current (f=2 kHz, V=14 kV_{pp}).

Figure 1 shows typical electrical characteristics (discharge current and voltage) of a discharge operating in the Townsend mode. The current I_d consists of a single pulse per half-period. Its shape is reproducible from one discharge to another. The gas voltage plateau is a characteristic of the Townsend discharge. An important feature is 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 between two discharges when the electric field is low enough to "trap" them in the gas volume, and thus is a clear signature of the volume memory effect from one discharge to the other.

The evolution of the first discharges also points out the importance of this memory effect after ignition (Figure 2). The first two discharges differ from the following ones showing that the initial discharge event depends on the previous ones and that a few discharges are necessary to reach the dynamic equilibrium in space and surface charge density. The first discharge is (always) filamentary, the second one is a mix of a Townsend discharge and micro-discharges, and the third one is entirely a Townsend one.

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 for excitation frequency higher than few kHz [5].

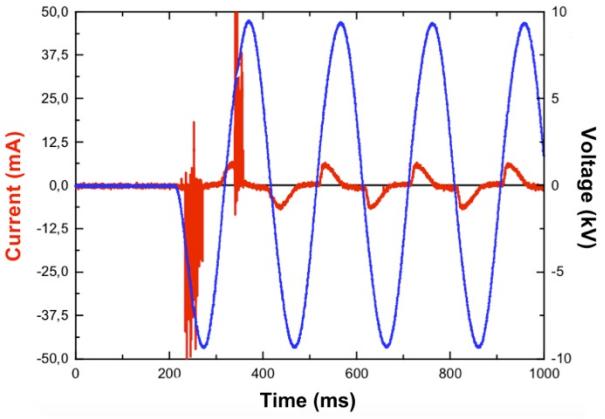

Fig. 2. Voltage and current waveforms of an APTD in N_2 during the first discharges.

4. Discussion

A source of seed electrons between two discharges is essential to enable uniform breakdown at low electric field strengths, i.e. obtain a Townsend discharge. This is referred to as the memory effect, which "slows down" the ionisation process during the gas breakdown and avoids streamer formation leading to a filamentary regime. Seed electrons between two successive discharges can come from the dielectric surfaces and the gas bulk.

Generally, the surface of the dielectric covering the anode is charged with electrons during the discharge. These electrons could be released to the gas gap when the anode becomes the cathode during the subsequent half-period. The mechanisms related to the surface charges can be separated into two categories. First, the secondary electron emission leads to the release of trapped electrons from the surfaces. It can be enhanced by collisions of metastable species such as $N_2(A^3\,\Sigma_u^+)$ with the dielectric surface [5]. These neutral species remain near the anode, where they are created during the discharge. According to the literature, the $N_2(A^3\,\Sigma_u^+)$ lifetime is a few tenth of µs for low concentrations of oxidising species [9]. Depending on the frequency, it may be shorter than the time between two discharges (roughly corresponding to 1/4th of the period). Thus, their density can remain high enough between two discharges to significantly impact the production of seed electrons for frequencies down to 2.5 kHz. However, the enhancement of the secondary electron emission by the collisions of $N_2(A^3\,\Sigma_u^+)$ with the dielectric surface(s) could be negligible for lower frequencies. Moreover, adding oxidising gas to nitrogen increases the quenching of the metastable species and reduces their contribution to the memory effect.

Second, the electrons trapped on the dielectric surfaces can also be released spontaneously into the gas gap [10]. This mechanism allows releasing seed electrons in the gas under a low electric field, independently of the gas composition and the applied voltage frequency. At very low frequencies, when the species in the gas bulk are completely renewed between two subsequent discharges, this mechanism prevails over the the memory effect. It can also play a significant role in air, as the quenching rate of the metastable nitrogen species such as $N_2(A^3\,\Sigma_u^+)$ is high. It depends on the nature of the dielectric surfaces [11] and their capacity to retain electrons [12-13]. Its contribution seems only predominant at very low frequencies and in other gases such as air, where no other memory effect mechanisms come into play [14].

The other type of mechanism responsible for the memory effect concerns the production of seed electrons in the gas bulk. Indeed, different chemical reactions occurring in the gas bulk can produce electrons. These mechanisms have been highlighted by adding small concentrations of oxidising gas in nitrogen [7,15]. The creation of seed electrons by the associative ionisation reaction between $O(^3P)$ and $N(^2P)$ is currently the most likely assumption that can explain the increase of the volume memory effect in such conditions [7-8]. The positive ions in the gas created by the associative ionisation reactions (for example, NO^+ ions produced by the reaction between $O(^3P)$ and $N(^2P)$) can also contribute to the memory effect, as was suggested by F. Massines [6] and Nemschokmichal *et al.* [16]. When the gas gap voltage increases just before the breakdown voltage, these positive ions increase the secondary electron emission at the cathode. This hypothesis requires further investigations to be confirmed.

The different mechanisms responsible for the memory effect in APTDs and their quantification depending on the

experimental conditions are summed up in Figure 3. In Figure 3(a), their respective contributions are compared as a function of the oxidising gas concentration, for a fixed frequency of applied high voltage of a few kHz. In Figure 3(b), they are compared as a function of the power supply frequency for gas mixture of nitrogen with a small concentration of oxidising gas (a few 10^{th} of ppm). It shows that the dominant physical and chemical mechanisms involved in the memory effect are strongly related to the experimental conditions (gas composition, dielectric material, operating HV, …).

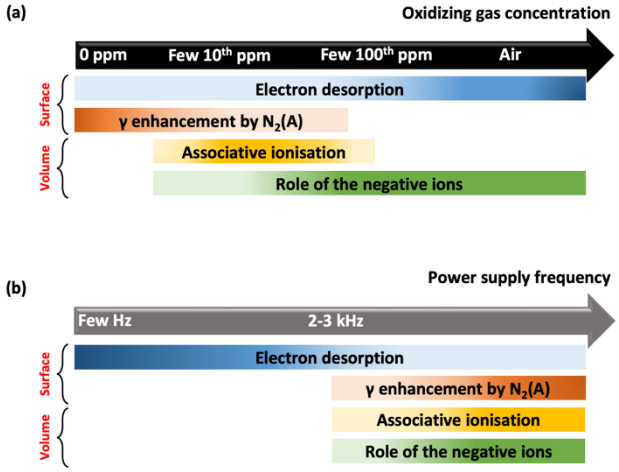

Fig. 3. Schematic representation of the dominant memory effect mechanisms as a function of: (a) the oxidising gas concentration at a fixed frequency of a few kHz, and (b) the frequency of applied high voltage (HV) in nitrogen with a small concentration of oxidizing gas.

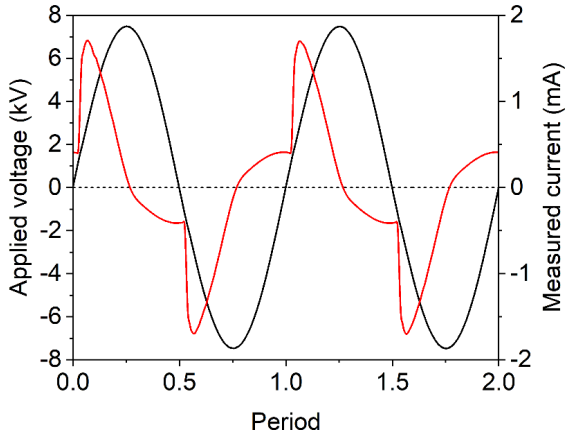

Fig. 4. Voltage and current waveforms of an APTD in CO_2.

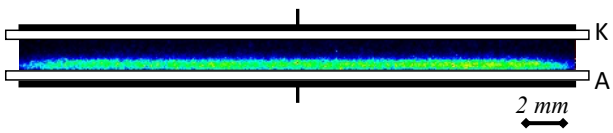

Fig. 5. Visualisation of the Townsend discharge in CO_2 using the iCCD camera with an exposure time equal to half of the period.

Therefore, understanding the predominant mechanisms depending on the experimental conditions allows a better control of the discharge stability, and the power dissipated in the discharge, for further development of plasma processes. For example, this understanding allowed us to understand how to obtain a diffuse discharge in other gases, such as pure CO_2 or N_2O. The figures 4 and 5 present respectively electrical measurement and picture of a discharge obtained in pure CO_2. Both are consistent with a Townsend discharge like in nitrogen. Indeed, the discharge current is composed of one single, long current peak for each half-period of the applied high-voltage, and the emission intensity is radially homogeneous and located near the anode side.

5. References

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