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Maximization of the working domain of an Atmospheric Pressure Townsend Discharge (APTD) using a current-source static converter

M.C. Bouzidi, X. Bonnin, N. Naudé, H. Piquet, A. Belinger, N. Gherardi

Université de Toulouse ; UPS, INPT ; LAPLACE (Laboratoire Plasma et Conversion d'Energie) ; 118 route de Narbonne, F-31062 Toulouse cedex 9, France

CNRS ; LAPLACE ; F-31062 Toulouse, France

nicolas.naude@laplace.univ-tlse.fr

Abstract. The objective of this paper is to present the implementation of a square-wave current inverter power supply, and to describe its effect on the working domain of an Atmospheric Pressure Townsend Discharge (APTD) in nitrogen. The results show that it allows a substantial increase of the percentage of time during which the discharge is ON (*on-time*) and therefore of the power delivered to the discharge, reaching almost 10 W/cm² in pure nitrogen. Moreover, the increase of the *on-time* of the discharge implies a decrease of the extinction *off-time* between two discharges, and thus an increase of the memory effect from one discharge to the following one, as it is monitored by using electrical and optical measurements. This leads to a larger working domain of the ATPD, even when adding impurities in nitrogen.

1. Introduction

Dielectric Barrier Discharge (DBD) is an easy and robust solution to generate low temperature discharge at atmospheric pressure. This discharge has a lot of applications (*e.g.* ozone generator, excimer lamp, plasma display panel (PDP), surface treatment, etc.) since more than one century [1-5]. Its configuration uses dielectric barriers to maintain current densities below the threshold for glow-to-arc transition. Depending on the gas, the electrical parameters, and the electrode configuration, dielectric barrier discharges (DBD) can operate in a homogeneous or filamentary mode [6]. Homogeneous DBD at atmospheric pressure have been obtained in helium, argon, nitrogen [7]. 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) [8].

A homogeneous DBD, especially if sustained in a low cost gas like nitrogen, is a promising alternative to replace some of the low-pressure plasma processes employed for thin film deposition [9]. The main expectations for this approach are to allow continuous processing and to reduce the equipment costs by avoiding the need for expensive pumping systems of conventional low-pressure plasma devices. However, the thin film growth rate using ATPD is still too low for an online process. Firstly, to improve this point, the discharge power must be increased [9]. However, the discharge



power is limited due to the fact that the slope of the voltage versus time is limited. Thus, in case of sinusoidal voltage power supply the amplitude and frequency of the applied voltage allowing to get the APTD are limited. A too high value of these parameters induces instabilities of the APTD which leads to a pure filamentary discharge. A solution to increase the discharge power is to control the current amplitude and to increase the discharge duration. This can be realized using a square-wave current inverter source [10]. Secondly, the reactive gases concentration in the discharge mixture has to be increased. Indeed, as can be seen on Figure 1, the thin film growth rate using APTD is limited not only by the discharge power but also by the precursor concentration. For example, Massines *et al.* [11] found out that using a classical sinusoidal voltage power supply, the Townsend regime can be successfully sustained only with hexamethyldisiloxane (HMDSO) dilutions in N_2 lower than 50 ppm. In case of O_2 admixture, the maximal rate has to be lower than 500ppm [12-15].

In this study, we report the use of a square-wave current converter in place of the classical sinusoidal voltage power supply to sustain an APTD in nitrogen. The materials and methods are described in section 2, and a discussion is done in section 3 concerning our motivations for the use of a square-wave current inverter. Sections 4 and 5 are dedicated to the results, focusing more specifically on the impact of the power supply nature on the discharge power (section 4) and on the discharge stability with respect to the additives (section 5).

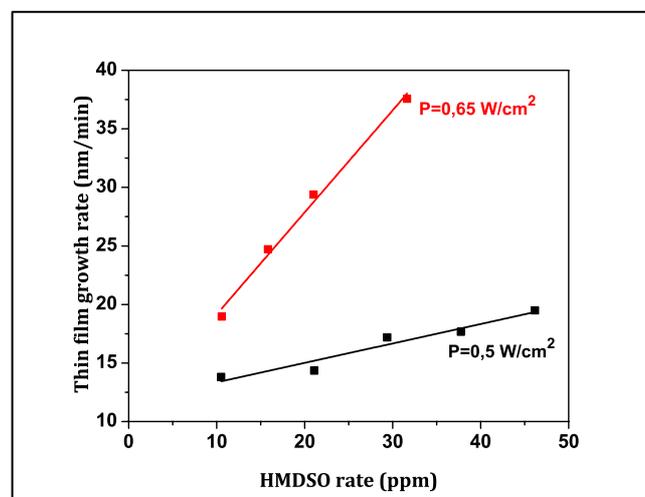


Figure 1. Effect of the HMDSO rate and discharge power on the SiO_x thin film growth rate using APTD in N_2 - N_2O -HMDSO mixture [16].

2. Experimental set-up

The DBD is kept in a closed vessel to perform experiments in a very well controlled atmosphere. The plasma reactor is pumped down to 10^{-3} mbar prior to any experiment, and then is filled up to atmospheric pressure using mixtures of nitrogen (99,999% purity) and synthetic air ($N_2/O_2=80/20\%$ with 99,999% purity) purchased from Air Liquide. The discharge is ignited between two alumina plates separated by a 1mm gas gap (Figure 2) and the discharge area is $3 \times 3 \text{ cm}^2$. In order to renew the atmosphere, a gas flow (1slm) 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 voltage power supply is made of a low-frequency generator providing the reference waveform, which is then amplified by a linear amplifier (Crest Audio model 8001, 4800VA) whose output is applied to the primary winding of a transformer (Montoux, ratio=150, 600VA, 60V/9kV) in series with a 4Ω resistor. The electrodes are connected to the secondary of the transformer. The discharge is characterized by electrical measurements. The discharge current is measured through a

200Ω resistor in series with the electrodes. The voltage applied to the electrodes is measured by means of a high voltage probe (Tektronix P6015A) whereas the voltage really applied to the gas, V_g , is calculated from the electrical measurements as described by Liu *et al.* [17]. The current and the voltage applied to the electrodes are visualized on a digital oscilloscope (Lecroy WaveRunner HRO 66Zi, bandwidth: 600MHz). The discharge homogeneity is investigated by means of short exposure time pictures, which are taken with an intensified CCD camera (Princeton Instrument PI-Max 3) synchronized with the power supply voltage.

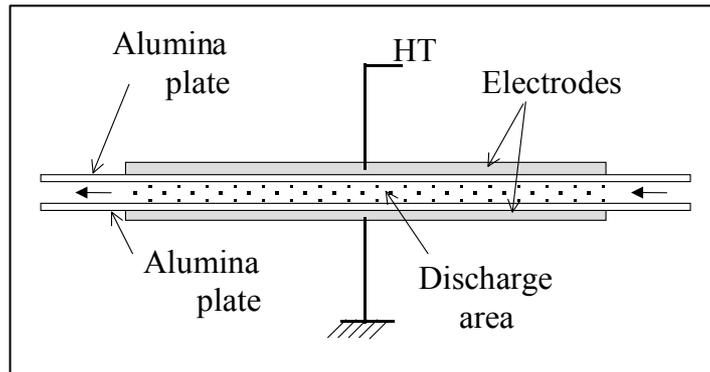


Figure 2. Schematic diagram of the discharge cell.

3. Which power supply in order to increase the discharge power keeping an Atmospheric Pressure Townsend Discharge?

Usually, APTD is obtained using a high-voltage low-frequency sinusoidal voltage power supply [8]. As already discussed, the amplitude and frequency of the applied voltage allowing to get the APTD are limited (Figure 3). The lower limit of the working domain, at a given frequency, is the applied voltage needed to extend the discharge over the whole electrode surface. And the upper limit, at a given frequency, is the applied voltage at which instabilities appear and so defines the transition to a filamentary discharge. This maximum voltage decreases when the frequency increases.

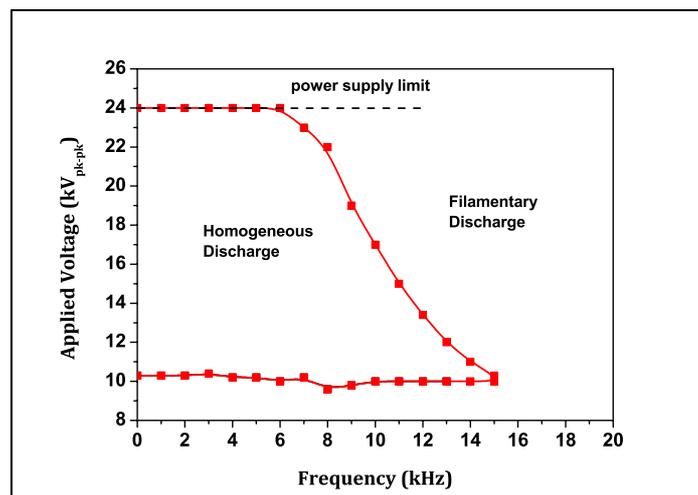


Figure 3. Working domain of the APTD in pure N_2 (the first point is obtained with $f=50\text{Hz}$).

In order to find out how to increase the discharge power in the homogeneous regime (APTD), it is interesting to take a look at the electrical measurements (Figure 4). The first point is that the maximal

discharge duration is about 60% of the period. The second point is that the APTD being a Townsend discharge, the gas voltage becomes almost constant after the discharge ignition. Thus, during the discharge, the voltage fluctuations are applied on the solid dielectrics and the current is derived from the solid dielectric capacity and the power supply voltage variation with time, *i.e.* the external circuit controls the discharge current [18]. Hence, the maximum limits of the frequency and amplitude of the applied voltage can be associated with a maximum current density allowing to obtain a homogeneous discharge: higher is the frequency or the amplitude of the applied voltage, higher is dV/dt , and so higher is the discharge current with the risk of reaching a too high ionization level to maintain a homogeneous discharge (transition to a filamentary discharge) [8].

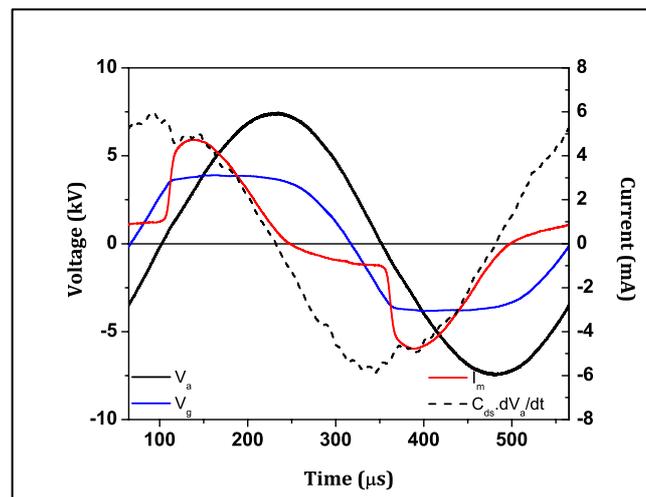


Figure 4. Oscillogram of the applied voltage V_a , the calculated gas voltage V_g , the measured current I_m and the current calculated from $C_{ds} \cdot dV_a/dt$.

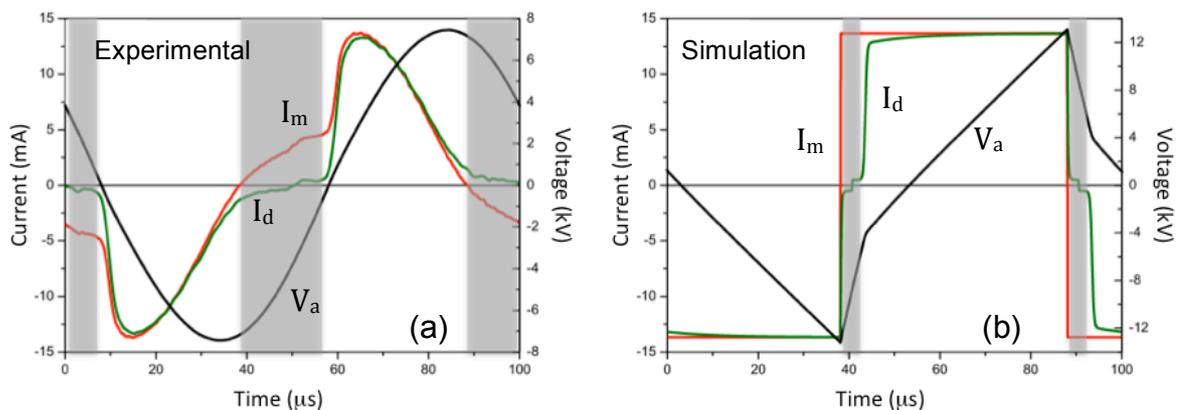


Figure 5. Comparison between sinusoidal voltage source (a) and square-wave current source (b) for the same discharge current amplitude: applied voltage (V_a), measured current (I_m) and discharge current (I_d) (the discharge is off in shaded areas) [9].

Therefore, to increase the discharge power a solution is to control the current and to increase not any more the discharge current amplitude but the discharge duration. This can be realized using a square-wave current inverter source [10]. The Figure 5 presents a comparison between experimental waveforms for a sinusoidal voltage source and a simulation of a square-wave current inverter source

for the same discharge current amplitude. The simulation is realized with PSpice software using a macroscopic electrical model of the discharge. This model treats the discharge as a Zener diode in series with a R-C parallel circuit. It takes into account i) the Townsend discharge breakdown, ii) the memory effect from one discharge to the following one and iii) the variation of the secondary emission coefficient during the breakdown [18]. In this case, the use of a square current waveform increases the duration of the discharge ignition from 64 to 90% and the power from 1.6 to 3.8W·cm⁻².

4. Comparison between sinusoidal voltage and square-wave current power supplies

The square-wave current power supply is a cascade structure composed of three parts: a tunable regulated DC current source, a current commutator to transform the DC current into an alternative square waveform current and a high voltage transformer. It has already been described in previous publication [10]. The Figure 6 shows experimental electrical waveforms obtained with this current power supply. As can be seen, the measured current is not perfectly rectangular. According to Bonnin *et al.* this phenomena is due to the parasitic elements (mainly the magnetizing inductance and parasitic capacitances) of the transformer [10]. Moreover, oscillations are observed on the current due to the switching of power converter. It is interesting to note that the capacitive nature of the discharge imposes a quasi-triangular applied voltage.

The homogeneity of the discharge is studied by short-exposure-time pictures taken with an intensified CCD camera. The Figure 7 shows a picture of the discharge during the positive half-wave with an exposure time of 100 ns. The light is maximal near the anode and homogeneous over the whole surface of the electrodes. This behavior is similar to what is observed in APTD using a sinusoidal voltage source [8]. As the discharge is homogeneous, it is possible to calculate the discharge current and the gas voltage (Figure 6). This allows to clearly determine the duration of the discharge (time for which the gas voltage is higher than the breakdown voltage, *i.e.* 3kV). Thus, it is interesting to note that differently from the conventional sinusoidal voltage case (Figure 4), the discharge is switched on during 85% of the period.

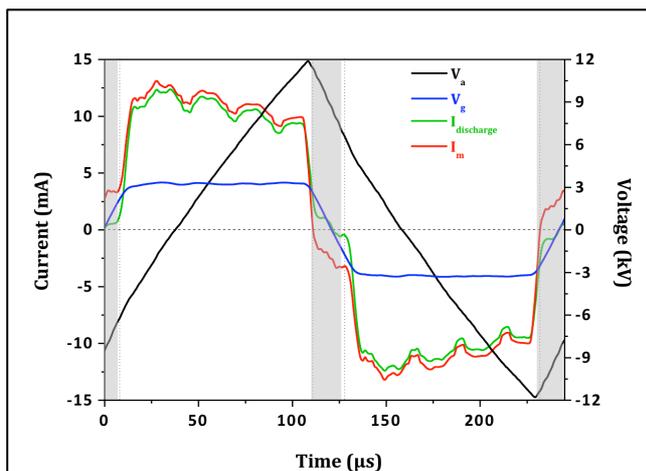


Figure 6. Oscilloscope of the applied voltage V_a , the calculated gas voltage V_g , the measured current I_m and the calculated discharge current (the discharge is off in shaded areas).

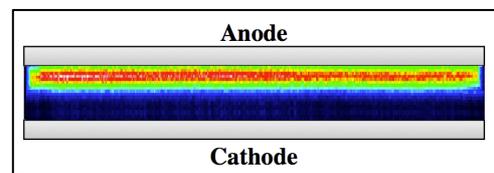


Figure 7. 100 nanosecond exposure time photograph of the gas gap using square-wave current inverter source.

The Figure 8 shows the evolution of the maximal power density according to the excitation frequency for the two types of power supply. The maximal power density is calculated at the frontier between the homogeneous and the filamentary discharge when it exists or for the power supply limit. Anyway, it corresponds to the upper limit of the working domain shown on Figure 3 in the sinusoidal

voltage case. At low frequency, the maximal insulation voltage of the cell discharge (24 kV_{pk-pk}) induces a limitation of the power density. This explains the linear growth of the power density for frequencies lower than 6kHz (sinusoidal voltage power supply) and 12kHz (square-wave current power supply).

As it can be clearly seen, the square-wave current power supply allows to increase the maximal power density by 79% up to 9.3W/cm². It is interesting to note that in case of the square-wave current power supply, the working domain *i.e.* the power density is always limited by the maximal insulation voltage (for frequencies lower than 12kHz) or the maximal current delivered by the DC current source (for frequencies higher than 12kHz). Hence, this indicates that it will be possible in a near future to reach values even beyond 10W/cm². In any case, at any given frequency, the power density is greater with the square-wave current power supply.

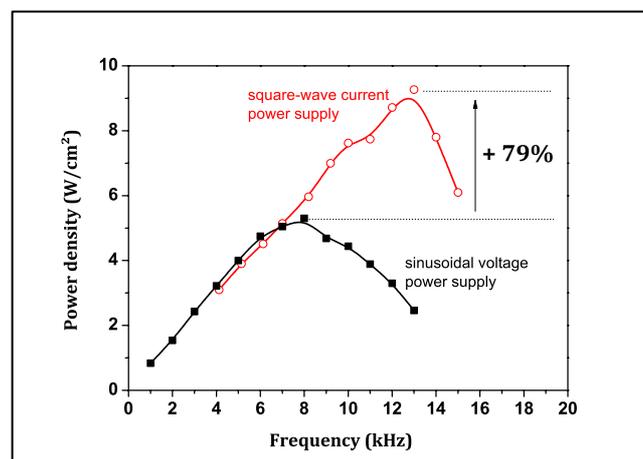


Figure 8. Influence of the power supply type (current or voltage) on the maximal discharge power according to the frequency.

5. Influence of the square-wave current inverter source on the stability of the APTD

This section discusses the influence the use of the current source could have on the stability of the discharge in the presence of additives. As discussed in the introduction, the APTD remains stable as long as the O₂ concentration is less than a few hundred of ppms. This effect is due to the quenching of N₂(A³Σ_u⁺) metastables state. Indeed, Massines *et al.* [8] found out that these long lifetime species are responsible for the memory effect from one discharge to the next, which is essential to obtain the Townsend regime at atmospheric pressure. The electrical signature of the memory effect is visible on the discharge current [15]. Indeed, this current is never equal to zero when the discharge is off (Figure 6) which is attributed to the continuous emission of electrons from the cathode induced by the metastable N₂(A³Σ_u⁺) flux [19].

As seen in the above discussion, the square-wave current power supply can increase the discharge ignition time and therefore reduce the time between two consecutive discharges (*off-time*). The Figure 9 presents the evolution of the *off-time* according to the excitation frequency respectively in the case of the current and voltage power supplies. Whatever the frequency, the *off-time* using the current power supply is at least two times smaller than the one using the voltage power supply. A reduction of this *off-time* implies a shorter time for the quenching of N₂(A³Σ_u⁺) metastables state in between two discharges. Therefore the memory effect from one discharge to the next must be higher, which is confirmed by looking to the discharge current amplitude before the breakdown (Figure 10): as it can be seen, for a given discharge power this current is higher with the square-wave current power supply than with the sinusoidal voltage power supply.

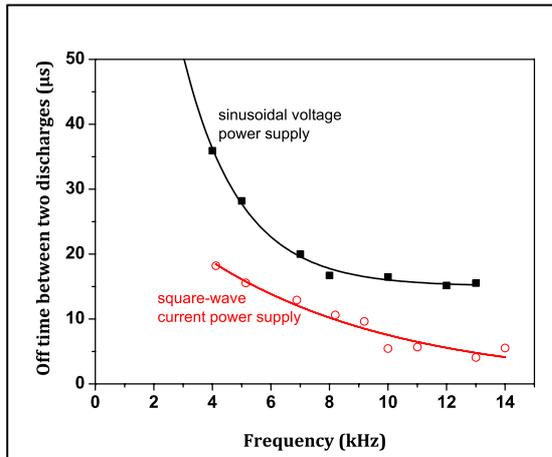


Figure 9. Evolution of the *off-time* between two discharges in the operating range limit for the two types of power supply.

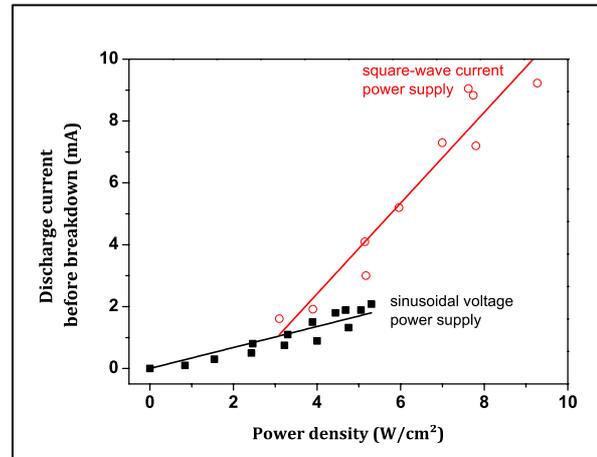


Figure 10. Influence of the power supply type on the electrical signature of the memory effect.

Moreover, the Figure 11 shows the evolution of the average power density for both power supplies as a function of the added O_2 rate to N_2 . The use of the square-wave current power supply allows keeping a constant power almost equal to 10 W/cm^2 up to 1000 ppm of O_2 , and an APTD can be sustained up to 1400 ppm of O_2 . On the other side, in the case of the voltage power supply, the average power density hugely depends on the O_2 rate: for 100 ppm of O_2 added to N_2 , the maximal power density increases a lot compared to pure N_2 , which has been already observed previously [14], and for higher concentrations, the power density decreases linearly down to 3.5 W/cm^2 for the maximum O_2 rate of 800 ppm in our conditions.

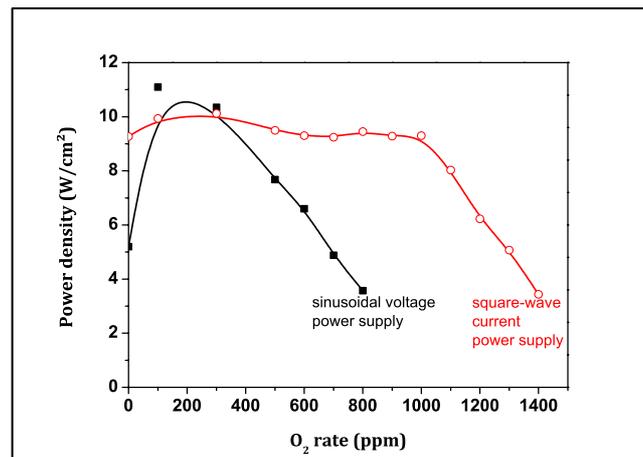


Figure 11. Evolution of the average power density at the limit of homogeneous domain as function of the rate of O_2 added to N_2 for the two types of power supply.

Then, by reducing the *off-time* between two discharges, the square-wave current power supply decreases strongly the sensitivity of the discharge regarding gas mixture. Thus, it is possible with this square-wave current power supply to increase both the discharge power and the additives concentration, which is very promising for thin film deposition processes.

6. Conclusion

In the case of an ATPD, the homogeneous discharge working domain is limited by a maximum density current. Moreover, the gas voltage is nearly constant during the discharge ignition, and so the discharge current is directly proportional to the variations of the applied voltage variations. As a result the use of a voltage source is not an ideal solution to supply the discharge: the APTD is turning off during a long time (nearly 40%), and the maximum current density is reached during a very short time. A square-wave current source appears more adapted: maximum current density could be reached almost all the period and therefore the discharge is turning off during a very short time. Consequently, the power density is higher with a current source: in this paper, the maximal power density is increased by 79 % with a current source while the used power supply is intrinsically limited. This suggests that it is still possible to increase the power to higher values.

Moreover, as the *off-time* is strongly reduced with the square-wave current power supply, the quenching of the $N_2(A^3\Sigma_u^+)$ metastable states in between two discharges is shorter. Then, the memory effect from one discharge to the following one is improved, which is verified in N_2/O_2 mixtures, the discharge staying in the APTD mode for the highest oxygen rates. Hence, for thin film deposition application, the use of a square-wave current power supply should allow to increase both the precursor concentration and discharge power, and thus the deposition rate.

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