A DIFFUSE NANOSECOND DISCHARGE FOR GENERATION OF REACTIVE SPIECES IN ATMOSPHERIC AIR

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ABSTRACT

A Non-Thermal Plasma (NTP) is created by a diffuse discharge generated under a nanosecond range overvoltage. The influence of the anode composition on the pattern of the discharge is studied and time-resolved image of the discharge developed under these conditions are presented. The focus is made on two different anode materials (copper and tungsten) leading to two distinct structures of the discharge: a well-defined and well-structured filamentary discharge and a filamentary-diffuse discharge with a glow around the anode extending until half-radius of the gap.

1. INTRODUCTION

In a Non-Thermal (or non-equilibrium) Plasma, the mean energy of electrons (few electron volts) is much greater than the kinetic temperature of ions and neutral species (less than few hundreds Kelvin). Energetic electrons produce free radicals, excited states species and ions trough dissociation, excitation and ionization of gas molecules by inelastic electron collisions rather than heating the global gas.

The most common way to create a NTP is by using gas discharges. When an electric field, with an amplitude above the breakdown field E_b, is applied between two electrodes, at a distance d away from each other, ionization process occur and lead to the formation of electron-ion pairs [1] and others species depending of the shape of the electron energy distribution function. The critical value of the breakdown field depends of both pressure p and distance d.

At low $p{\times}d$, the gas discharge formed is called a glow discharge and creates a large volume of NTP. But as $p{\times}d$ increases the current density increases and the discharge constricts until forms a thin conductive channel. The resulting plasma is inhomogeneous in the inter-electrode space. If the current density continues to increase then discharge transits to an arc discharge and forms a thermal (or equilibrium) plasma, in which all temperatures (electron, ions, neutral species) are equal [2].

At atmospheric pressure, to produce a NTP, the current density growth must be constricted either by limiting the distance d (few millimetres) or by cutting the electric field. But in general the volume of plasma created by the discharge is small. To create a large volume (more the few cm^3) of plasma one can modify different key parameters [3]: electrode configurations, discharge excitation modes, or external ionisation systems (photo-triggered discharge, electron-beam). Atmospheric pressure NTP might be use in numerous applications especially in environmental applications [4].

Since 1970, an important work has been dedicated to the study and comparison of different kinds of NTP, such as DBD, surface, preionised [5], corona or nanosecond pulsed discharge in the scope of removal of Volatile Organic Compound (VOC) from gas flows, [6]. Each plasmas created is more or less efficient to remove VOCs, according to the type of molecules to be treated, to the time and space energy deposition and to the volume of plasma that can be generated. Globally it has been shown that more the energy deposition is homogeneous and better is the treatment efficiency. The challenging task is to create such large volume of homogeneous NTP at atmospheric pressure without preionisation.
Recent experimental studies in pin-to-plane geometry have shown that a large volume of NTP could be induced by high-pressure diffuse

discharges under very strong overvoltages, higher than 17 kV/ns [7]. In the present study, using this electric excitation, the dynamic of the discharge in atmospheric air is investigated for a coaxial wire-to-cylinder geometry. Most particularly, the effects of the anode wire composition (copper or tungsten) are presented. Times resolved imaging and fast electrical measurements are used to characterize the discharge.

2. EXPERIMENTAL SET-UP

The discharge is created in a reactor composed of a copper cylinder (cathode) and a thin wire of tungsten or copper (anode) with a diameter of 50 µm for a total length of 27 mm. The inter-electrode distance d is 13 mm, for a reactor volume of 14.280 cm^3. A single pulse of a positive high voltage (up to 80 kV) is delivered to the wire by a homemade generator.

The generator is based on a two stages Marx-type power supply coupled to a 50 Ω coaxial line through a spark gap. A Thyratron device triggers the first Marx stage. The two stages lines supplies a 40 kV pulse with a steep rise time of about 3-4 ns controlled by the synchronization of the spark gap commutation of each stage. Because of the high impedance of the discharge the pulse doubles and propagates backwards when reaching the end of the coaxial line. The reflected pulse arrives to a spark gap that commutes for a double amplitude signal and induces a shortcut of the coaxial line. Thus the voltage output drops to zero within 2-3 ns. This system can give a square shape to the pulse. The length of the coaxial line, L, and the signal velocity, $c=2.10^{10}\ cm.s^{-1}$ in the 50 Ω coaxial line, set the pulse duration, $T=2L/c$. For the experiments presented in this study, the duration at the full-width and half-height is set to 10 ns, with maximum amplitude around 80 kV and a rise-time of about 20 kV.ns^{-1}.

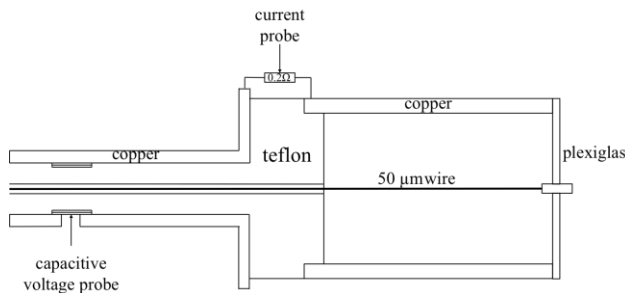*Fig. 1 Reactor scheme*

The cathode is grounded through a low inductance coaxial resistive shunt of 0.2 Ω. Thus, high-resolution current measurement can be done without electromagnetic disturbances. The voltage is recorded very closed to the reactor by a homemade capacitive coaxial probe around the coaxial line. Such integrated probes allow doing measurements without disturbing the discharge behaviour and losing information (Fig. 1). Discharge energy is derived from current and voltage measurements (Fig. 2). All the signals are recorded on a 2 GHz bandwidth digital oscilloscope with a 10 Gs.s^{-1} sampling frequency.

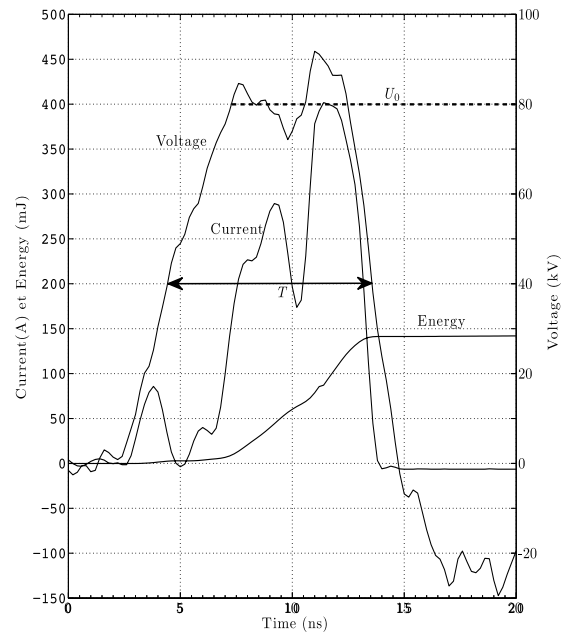*Fig. 2 Electrical records (U$_0$ = 80 kV, T = 10 ns)*

The discharge imaging is done with a 12 bits resolution Atmel CCD camera coupled to a pulsed fragment intensifier and fitted out with a 50 mm F/1:1.2 lens. The depth of field of the whole system is estimated to about 900 µm and the discharge imaging is made along the cylinder axis on a radial plane inside the cylinder. The intensifier gate can be set from 5 ns to 20 ns and can be properly synchronised with the voltage pulse, so that the development of the discharge can be resolved with a nanosecond resolution.

3. STRUCTURE OF THE DISCHARGE UNDER VERY STRONG OVERVOLTAGE

When a high voltage is applied to a gap in a very short period of a few nanoseconds, the breakdown can occur at levels much higher than

the static breakdown threshold. Applied to a very thin anode, this overvoltage leads to an extremely high electric filed, which could translate the electron energy distribution function towards higher values. The coaxial wire-cylinder geometry has high asymmetric electrodes, thus such levels could be reached.

On discharge pictures (Figs 3 & 4), the dark circular dashed circular line and the six small white circles along this line correspond to luminous artefacts created by furrow and holes on the optical window. The white spot in the middle is the non-transparent holder of the anode, corresponding to a blind spot of the discharge observations. The pictures (Fig. 3 & Fig. 4) are all synchronized with the beginning of the voltage rise; numbers on each of them correspond to the integration time of the intensifier. Rescaling the grey-scale between the max and min grey-values has made post-acquisition enhancements of the contrast. For each development stage, different light-gains have been used, thus stage comparison is only made on the structure evolution of the discharge development.
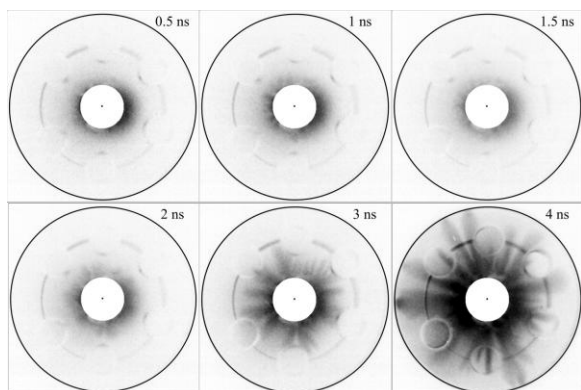

Fig. 3 Time resolved imaging of the tungsten-wire discharge (U_O=80 kV; T = 10 ns).

Based on the time resolved optical characterization, different stages for the development of the discharge could be defined: first a diffuse pattern that extends symmetrically from the anode and reaches around half-radius of the gap in about 2 ns. Then several secondary blurry channels emerge from the outer limit of the primary diffuse glow and propagate through the rest of the gap and reach the cathode about 2 ns later. After the cathode junction the spatial structure of the discharge does not change. This junction correspond to the steep rise of the current, up to 400 A, on the electrical records

and leads to a total energy of 140 mJ. Because of the voltage is cut-off very quickly after the current maximum the discharge does not transit to an arc.

For a copper-anode, the sequence of the discharge development does not change but the timing is different (Fig. 4). The first stage of the discharge development is 2 ns longer and the diffuse part is much more constricted around the anode. Thus the multi-filaments structure is created much closer to the wire. In this second stage of the discharge, filaments are thinner and more distinctive from each other than in the case of the tungsten-wire. Looking at the overall development time from the beginning of the HV-pulse to the cathode junction, it is 3 ns longer in the case of a copper-anode than for a tungsten one.
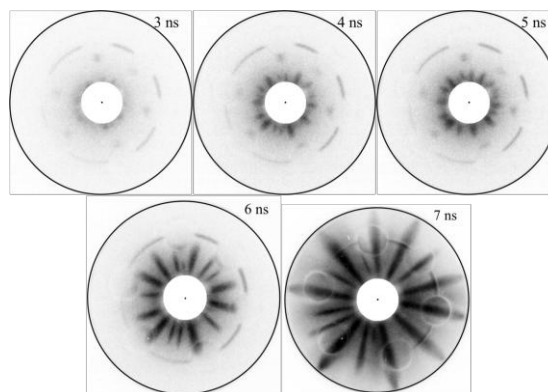

Fig. 4 Time resolved imaging of the copper-wire discharge (U_O = 80 kV; T = 10 ns).

In Fig. 5, the contrast of integrated pictures of the discharge has been enhanced by a factor 1.5. Intensity comparison is relevant since the light-gain is the same for these two pictures. It can be observed that the tungsten-wire discharge (Fig 5.a) is brighter and wider than the one for a copper-wire (Fig 5.b). The filamentary stage leads to a well-organised pattern with filaments more equally spaced out in the case of the copper-wire. On the other hand, the diffuse pattern is more distinguishable for the tungsten-wire but the space localization of the filamentary parts seems to be more random.

For both configurations, discharge currents can rise up to around 400 A. However, for the copper-wire, it leads to a slightly lower total energy of about 100 mJ because of the total duration of the current, which is about 3-4 ns for

the copper-wire, i.e. 2 ns lower than for the tungsten-wire. For the same voltage pulse characteristics, less energy can be deposited in the gaseous volume for the copper-wire configuration. Calculation of the discharge impedance at the current maxima gives a 200Ω equivalent resistance for both configurations.

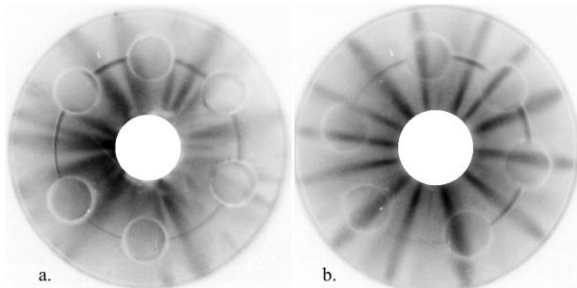*Figure 5. 15 ns integrated pictures of the discharge ($U_O = 80\ kV$; T = 9 ns): (a) tungsten-wire; (b) copper-wire*

4. CONCLUSION

A large volume of NTP plasma could be created in atmospheric air by a diffuse discharge under a nanosecond steep rise-front overvoltage. For a tungsten-anode, the discharge development becomes a combination of filamentary regime and a diffuse one, spreading on a quit large volume of about three quarters of the reactor volume (Fig 5.a). At its maximum extension, the diffuse pattern spreads over half of the reactor radius and then turns into diffuse plumes with no-space organisation with a propagation velocity of 3.25×10^6 m.s^{-1}. Whereas, with the copper-wire, the discharge has a filamentary structure from the early stage and shows a well-spaced and organized structure, representing a smaller volume of NTP. These differences could be linked to the hypothesis given in [7], for a point-to-plane air atmospheric discharge under the same kind of overvoltage. When high-energy electrons accelerated by the very high electric field collide with the wire, it could produce X-ray emission, which could pre-ionize the gas and promote a homogenous development of the space charge. In this case, the composition of the wire could be a key factor, which could determine the energy of this radiation and the depth of pre-ionization towards the cathode just as the value of the overvoltage. The discharge pattern observed with the tungsten-wire gives the larger volume of plasma. For pollutants removal applications, it means a larger volume of reactive species, which could participate somehow to the fragmentation of molecules. Thus, spectroscopy diagnostics and ozone measurements will be conducted on the diffuse discharge in dry air to quantify the production of radical oxygen and nitrous metastables.

REFERENCES

[1] Y. P. Raizer, "Gas Discharge Physics", Berlin Germany: Springer-Verlag, 1991.

[2] A. Fridman, A. Chirokov, A. Gustol, "Non-thermal atmospheric pressure discharges", J. Phys. D: Appl. Phys., **38**, R1-R24, 2005.

[3] E. E. Kunhardt, "Generation of a large-volume, atmospheric pressure, non-equilibrium plasmas", IEEE Trans., **28**, 189-200, 2000.

[4] T. Hammer, "Application of Plasma Technology in Environmental Techniques", Contrib. Plasma Phys., **39**, 5, 441-462, 1999.

[5] B. Lacour, V. Puech and S. Pasquiers,"High-pressure phototriggered discharges : Physics and applications", Recent Res. Devel. Applied Phys., **6**, 149-191, 2003.

[6] A.M. Vandenbroucke, R. Morent, N. De Geyter, C. Leys, "Non-thermal plasmas for non-catalytic and catalityc VOC abatement", Journal of Hazardous Materials, **195**, 30-54, 2011.

[7] P. Tardiveau, N. Moreau, S. Bentaleb and al, "Diffuse mode and diffuse-to-filamentary transition in high pressure nanosecond scale corona discharge under high voltage", J. Phys. D: Appl. Phys., **42**, 175202, 2009.