MODULATION OF IONIZATION FRONT PROPAGATION VELOCITY IN A µS PLASMA GUN HELIUM DISCHARGE WITH NITROGEN ADMIXTURE

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ABSTRACT

The propagation velocity modification of µs helium plasma gun (PG) discharge with nitrogen gas admixture in the helium buffer is studied in relation with ionization front imaging and helium-nitrogen plasma features. From 0,1 % to 0,8 % of N2, the ionization front is faster, more extended and with strong luminous intensity, compared to the high purity helium. The velocity has an optimum, for approximately 0,3 % of N2. Above 0,3 %, the velocity is progressively slowing down, and finally gets slower than the pure helium case above 0,9 %. The velocity evolution may be linked with kinetic and energy transfer predominantly on N2+ or N2 according to the increase of N2 %. Consequently, a local reinforcement of electric field in the front of the discharge is suspected for low N2 percentage, involving velocity increase of the discharge.

1. INTRODUCTION

Atmospheric pressure plasma jet devices have great potentialities for biomedical applications. In these devices, the plasma propagation relies on ionization wave generation with intense local electrical field in the ionization front. The front propagated inside capillary and finally outside of it (plasma plume). A lot of studies have been carried to understand this complex mechanism [1]. However, there still remains a lot of misunderstanding on the deep nature of these phenomena, especially because they combine various physical parameters.

One of them is the contribution of kinetic reactions and associate energy transfers. In this present work, we propose to evidence the pertinent kinetic reactions influence on the discharge development.

Experimentally, we proceed with N2 admixture for constant operating parameters (applied voltage, gas flow rate…). This led to isolate the relative role of kinetic reactions and evidence the associated important energy transfers in the discharge development [2].

In addition for biomedical applications, it could be an effective way to maximize the efficiency of plasma treatment. For example, very small amount of N2 in the helium buffer leads to significant increase discharge propagation distance along long capillary and could lead to plasma plume expansion.

The plasma source used in this work is a µs pulse powered driven PG [3]. The PG is a coaxial dielectric barrier discharge reactor with a quartz capillary, flushed with helium and powered by µs voltage pulses in the kHz regime. PG discharge generates an ionization front (and the ionized channel tail behind) that we called PAPS for Pulse Atmospheric Pressure Plasma Streams [3].

2. EXPERIMENTAL SETUP

The figure 1 shows the experimental setup.

A 40 cm long dielectric quartz capillary with a 4 mm inner diameter and 6 mm outer diameter is used. The inner electrode, 2cm long, is set inside the capillary. The helium buffer is injected through in the inner hollowed electrode (0,8 mm inner diameter). The nitrogen is mixed in the helium buffer.

Ten optical fibers are regularly disposed along the capillary, 2cm apart of each other, and connected to a photomultiplier tube (PMT). This setup allows to follow the plasma front displacement during a single discharge event, but also to measure its velocity on the distance between two fibers. Velocity calculated between two consecutive fibers will referred as

"instantaneous velocity". Velocity calculated between more spaced fibers (15cm for example), will referred as "average velocity".

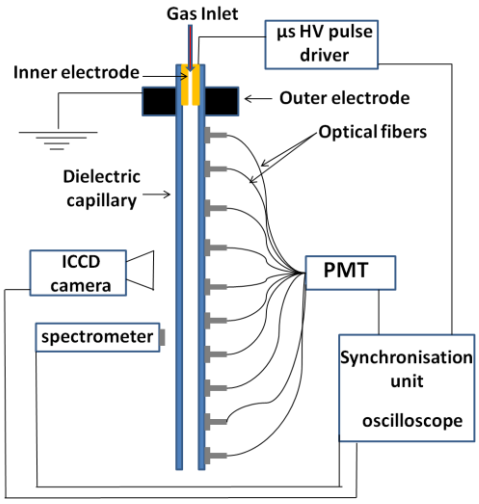

Fig.1: experimental setup

A PIMAX 3 fast ICCD camera is used to image the discharge.

Time integrated (200ms) emission spectra of the discharge are obtained by a 2000 maya pro spectrometer.

3. **RESULTS**

Experiments have been first carried out in helium without N2 admixture (pure helium) at a voltage of 16kV and a pulse repetition rate (prr) of 500 Hz.
Figure 2 shows the voltage time evolution together with the ten fiber PMT signal. The associated PAPS velocity profile along the capillary is shown in figure 3.
The PAPS front reaches the first fiber a little bit later than 1 µs after the pulse ignition. At this time, the applied voltage is approximately only 8 kV. 1 µs later, the pulse amplitude reaches it maximum (fig.2), and the velocity is higher and stabilized (close to 1, 3 x 10^7 cm/s in fig.3).

For other voltage amplitude (not shown here), the behavior is the same, with a shift of the velocity profile, depending on the time delay between the onset of the voltage pulse and the discharge ignition itself.
The voltage pulse shape time evolution has obviously here a dominant effect on the velocity profile. But the ionization front is associated with long time evolution ionized channel behind it. Consequently, charge deposition on the capillary walls could also modify the propagation velocity too (hysteresis phenomena). This fact has been evidenced in other plasma jet configurations [4].

In the following, all the results presented have been obtained with same discharge parameters: 16 kV and 500 Hz prr. Since the instantaneous velocities have very little variations between 8 and 20 cm along the capillary, we can take single average velocity between the fibers 3 and 10.

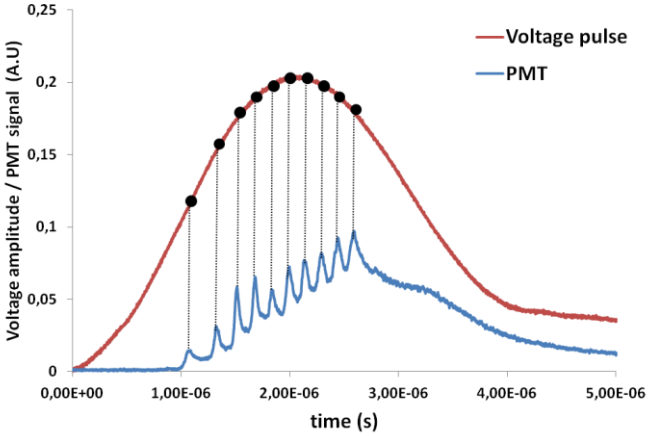

Fig. 2 : 16 kV applied voltage time profile synchronized with the PMT signal of ten optical fibers. The first fiber is located 3cm downstream from the inner electrode tip.

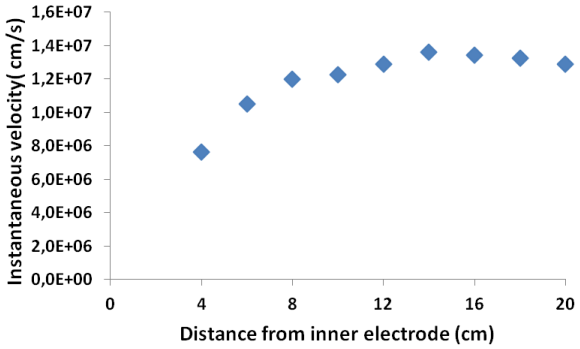

Fig 3: evolution of the velocity profile of the discharge inside the capillary. The velocity is calculated between two consecutive 2 cm spaced fibers.

Figure 4 shows the evolution of the average velocity of the PAPS versus nitrogen admixture.
It can be clearly seen that with only 0,1 % of N2, the PAPS velocity is significantly increased compare to pure helium case. The discharge velocity reaches its maximum around 0,25 %. Then for higher N2 admixtures, the velocity starts to decrease. At 0,9 % of N2, the discharge

velocity is close to the one in the pure helium case, and clearly lower for higher percentages.

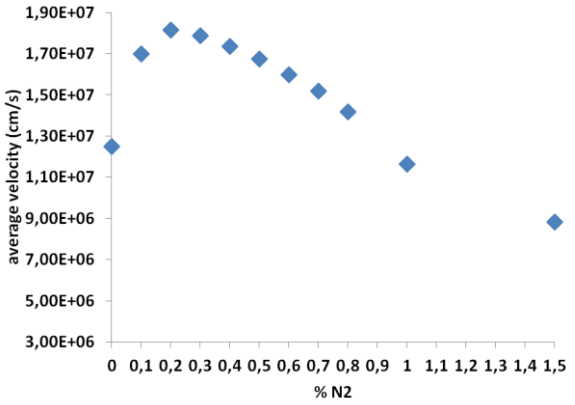

Fig. 4 : average velocity of the discharge with respect for N2 %. The average velocity is calculated here between the fiber 3 and 10.

Figure 5 shows the evolution of the PAPS ionization front propagating inside the capillary. Velocity measurements in fig.4 are correlated with fast ICCD images (set 15 cm after inner electrode)

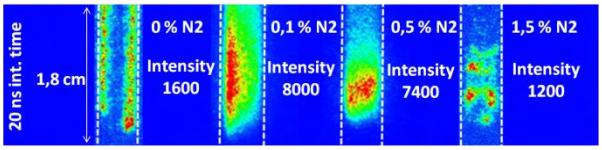

Fig. 5: ICCD images of the discharge inside the capillary (20 ns integration time) for pure helium case and 3 different % of N2. Same false color scale is used for intensity comparison (more intense in red).

In pure helium case, the pattern exhibits two plasma streams propagating on the capillary walls. Observations on this pattern and its strong dependence on the applied voltage and pulse repetition rate is discussed in [5]. At 0,1 % N2, we observe a drastic evolution of the shape of the ionization front. Indeed, the light is more homogeneous and intense with a single larger ionization front. One can note that the most intense area is still localized close to capillary wall. In relation with fig.4, this pattern of ionization front is correlated with the brutal velocity increase compare to pure helium case.
At 0,5 % N2, the ionization front has about the same intensity level than in the previous case, the PAPS head appears reduced homogenous and occupying the whole crossed section inside the capillary. We note that at this precise N2 %, the velocity is close to one with 0,1 % of N2 in fig.4, but the ionization front pattern is not the same. However, a gradual modification of plasma pattern ionization with N2 admixture increase is observed. When the N2 % is enough, the velocity is lower than pure helium case and the change in ionization front is strong. As we can see for 1,5 % N2 ,the front is fragmented, with several punctual intense luminous areas. We note here the light is strongly reduced compared to the previous case.

Figure 6 shows emission spectra of the discharge for different N2 admixtures (set 15 cm after inner electrode).

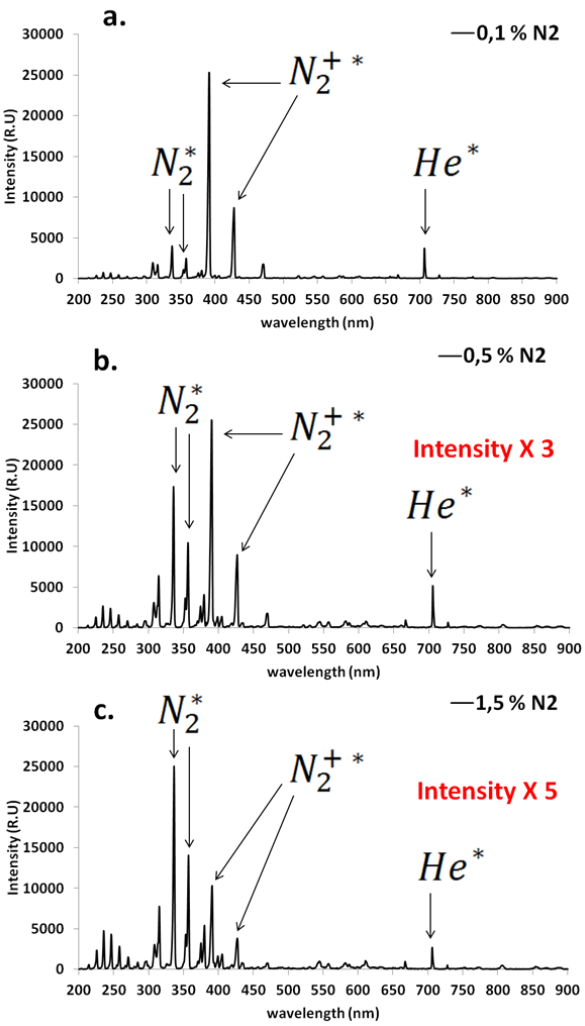

Fig. 6 : evolution of integrated (200 ms) discharge spectra through the capillary for 3 N2 % (a : 0,1%, b: 0,5%,c:1,5%)

In (a) (0,1 % N2) the N2+ band is very intense compared to all other excited species emission. When the % of N2 is increased to 0,5% (b) the emission level of N2 bands are close and not negligible anymore compared to N2+ bands. Finally at 1,5 % of N2 (c), 337,1 nm N2 band is the dominant one in the spectrum.
Concerning 706.5 nm He transition in the three spectra, it decreases when the N2 % increases.

Between 200 nm and 300nm, several bands of NO γ system appear. The higher the % of N2 is, the higher the NO γ bands emission is.

As a result, it appears that the intense light in the ionization front at 0,1 % in fig.5 is mainly due to N2+ emission. At 0,5 % of N2, the intensity of 391,4 nm N2+ is weaker compared to the previous case (25 000 vs. 8000), but the global intensity is more equally distributed between N2 and N2+ bands.

The relative changes between N2+/N2 bands intensity are the signature of energy transfer modification, preferentially to N2+ or N2 species with respect of the % of N2 admixture. In atmospheric pressure helium discharge, the charge transfer from He2+ and pening ionization He(2.3S) metastable [6] are very effective to form N2+ ions and keep relatively high ionization degree. If the concentration of N2 in the helium discharge is too high (above 0,5%), more energy is dissipated in the ro-vibrational levels of excited N2 species, leading to lower ionization degree.

The consequence of these modifications is suspected to impact on electronic temperature and density. For small percentage of N2 in the helium discharge, the local electrical field in the ionization front is strengthened by massive production of N2+, which is translated into strong velocity increase and pattern modification of the front shown in fig.4 and 5 and illustrated in 0,1 % case. Then at higher % of N2, the energy is progressively transferred to N2 ro vibrational levels at the cost of electronic temperature and ionization decrease. It results on velocity decrease and patterns modifications of ionization front as seen in fig.4 and 5 in the 1,5 % of N2 case.

Other mixing configuration (not reported here), where N2 is mixed downstream after the discharge reactor with specific T-shape quartz capillary exhibits similar velocity profiles with respect to % of N2 admixture. This observation minors phenomena at the discharge level and is consistent with the kinetic reactions mentioned above.

4. **CONCLUSION**

In this work, the study of nitrogen admixtures on PG helium plasma features, ionization front pattern and velocity, plasma emission is documented. The admixture of a few % is associated with the generation of intense ionization front travelling at the higher velocity in comparison with pure helium baseline case. Over this admixture range, the plasma emission gradually switch from a N2+ dominated to a balanced N2+/N2(C) and NO γ spectrum. For admixture higher than 1 %, ionization front pattern exhibit hot spots, significant velocity drop is measured together with a N2 dominated emission spectrum.

Such kinetic transfer in atmospheric pressure helium/ nitrogen plasma were already largely documented in relation with nitrogen admixture level and are correlated with plasma density and electron temperature dependent reaction rates. Beyond the modulation of reactive species and plume expansion for biomedical applications, this work compiled a data set which may constitute a valuable test bench for PAPS propagation modeling. Modeling of plasma propagation in helium/nitrogen could be validated with the comparison with plasma velocity and emission spectra. Invaluable but experimentally difficult to measure data such as reduced electric field in the ionization front, space charge and electron density and temperature would then be inferred from modeling.

Acknowledgment

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