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Experimental investigations of plasma bullets

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Abstract
Recently several investigators reported on various means of generating cold plasma jets at atmospheric pressure. More interestingly, these jets turned out to be not continuous plasmas but trains of small high velocity plasma packets/bullets. However, until now little is known of the nature of these 'bullets'. Here we present experimental insights into the physical and chemical characteristics of bullets. We show that their time of initiation, their velocity and the distance they travel are directly dependent on the value of the applied voltage. We also show that these bullets can be controlled by the application of an external electric field. Using an intensified charge coupled device camera we report on their geometrical shape, which was revealed to be 'donut' shaped, therefore giving an indication that solitary surface ionization waves may be responsible for the creation of these bullets. In addition, using emission spectroscopy, we follow the evolution of various species along the trajectory of the bullets, in this way correlating the bullet propagation with the evolution of their chemical activity.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The plasma jet used in this study was produced by a pulsed plasma source, 'the plasma pencil', that we developed and reported on previously [1]. Briefly, the plasma pencil is made of a 2.5 cm diameter hollow dielectric tube with two disc electrodes about the same diameter as the tube inserted into it and separated by a 0.5 cm gap. Each of the electrodes is composed of a thin copper ring attached to the surface of a centrally perforated dielectric disc [1]. To ignite the plasma, unipolar nanoseconds square high voltage (1–10 kV) pulses at repetition rates in the 1–10 kHz range are applied between the two electrodes and a gas mixture (such as helium and oxygen) is passed through the holes of the electrodes at flow rates ranging from 1 to 10 L min^{-1}. When a discharge is ignited in the gap between the electrodes (discharge current pulses reach magnitudes up to the ampere range but for only a few nanoseconds), a plasma plume reaching lengths up to 5 cm or more is launched through the hole of the outer electrode and in the surrounding environment [1]. The length of the plume/jet depends on the magnitude of the applied voltage, the gas type and its flow rate.

To better understand the dynamics of the plasma plume, a high-speed intensified charge coupled device (ICCD) camera with exposure times of a few nanoseconds was used to capture the temporal emission. It was found that the plasma plume is not a continuous plasma but rather a moving plasma bullet [2]. Similar observations were also made by other investigators with other devices [3, 4]. The bullet starts to accelerate as soon as it is launched from the hole of the outer electrode. At some distance it reaches its maximum velocity and then the velocity drops quickly until the bullet cannot be captured by the ICCD anymore. The maximum value of the plasma bullet velocity is in the order of 10^5 m s^{-1}, a much greater value than the gas velocity of 8.3 m s^{-1}, as calculated according to the gas flow rate and the diameter of the hole. To explain the high propagation velocity of the plasma bullet under a very weak electric field condition, Lu and Laroussi [2] invoked a photo-ionization based model. In this model, the head of a cathode-directed streamer is a sphere of radius r_0, containing n^+ positive ions. As it moves forward, it

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leaves behind a quasi-neutral ionized channel with a negligibly low conductivity; the head is not connected to the anode and only the streamer head is measurably luminous. This corresponds to what was observed experimentally. Because of emission of photons from the streamer, photoelectrons are created at a suitable distance r_1 from the centre of the sphere. Under the influence of the field set up by the space charge, the electrons are accelerated towards the sphere and an avalanche is initiated. If the multiplication up to the sphere is sufficient, the electrons neutralize the positive charge, but leave behind a new positive region. Photons emitted from this region create photoelectrons at a new suitable distance and the process goes on until there is not enough energy to sustain the ionization of the new gas volume replaced by the streamer channel [2]. According to this model, the plume velocity can reach values as high as 10^6 m s^{-1} and it can travel up to several centimetres without the presence of an externally applied electric field. This is in agreement with experimental observations.

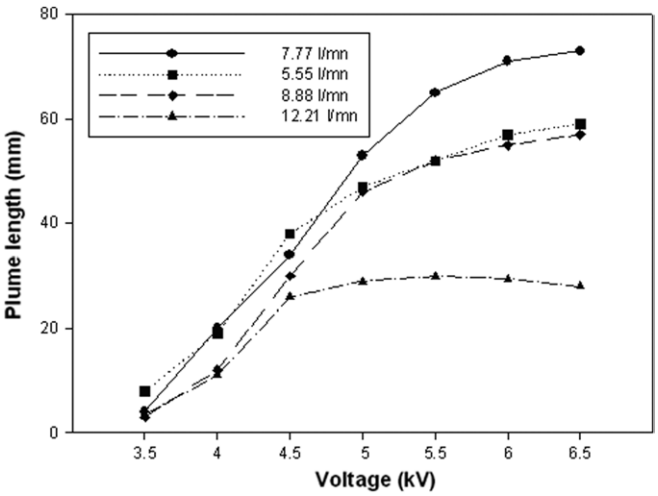

Figure 1. Plume length versus voltage for different helium flow rates. The voltage pulse width is 800 ns.

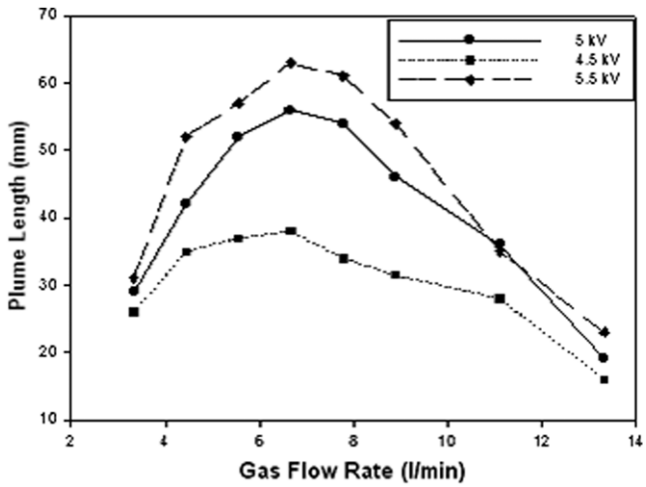

Figure 2. Plume length versus flow rate for different voltages. The voltage pulse width is 800 ns.

In this paper, to further understand and characterize the plasma bullets, ICCD imaging and spatially resolved emission spectroscopy are used to investigate the propagation of the bullet, reveal its structure and follow the evolution of its important chemical species, such as O, N_2, N_2^+ and OH. The parameters influencing the characteristics of the bullet are identified, leading to some rather surprising results and pointing the way to the possible physical processes responsible for the creation of these bullets. In addition, in this paper we report for the first time a new and very interesting observation: applying an external dc electric field to the plume impacts both the length of the plume (distance travelled by the bullets) and the time of initiation of the plasma bullets. Therefore, we introduce here a means by which to control the dynamics of the bullets. This is studied by using an ICCD (DiCAM-PRO) and measuring the dependence of the velocity of the bullets with the magnitude of the externally applied dc voltage.

2. Plume length investigation

The length of the plume created by the plasma pencil depends mainly on two parameters: the applied voltage between the two electrodes and the helium gas flow. As is the case for most other plasma jets the plume is electrically driven; however, the gas flow appears to have an influence on the length and the shape of the plume. This is due to the creation of a helium gas channel through the surrounding air within which an ionization front is allowed to propagate forward. For optimum operation a proper ratio between the size of the hole and the flow rate has to be found in order to get a laminar flow and avoid turbulence. In this case, the gas is well channelled and prevents mixing with the ambient air for a longer range.

The voltage is the main parameter affecting the length of the plume. Figure 1 shows the influence of the voltage on

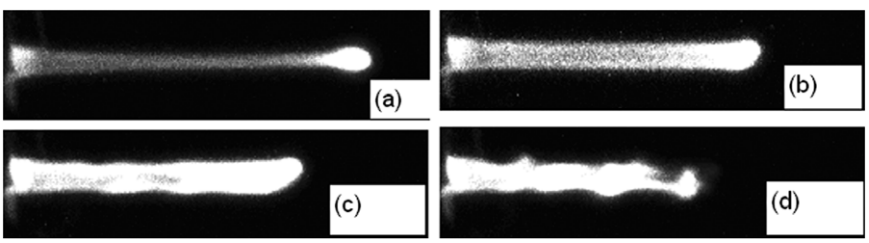

Figure 3. Shape of the plume for different flow rates: (*a*) 6.6 L min^{-1}, (*b*) 8.8 L min^{-1}, (*c*) 11 L min^{-1} and (*d*) 13.2 L min^{-1}. Applied voltage is $V = 5$ kV.

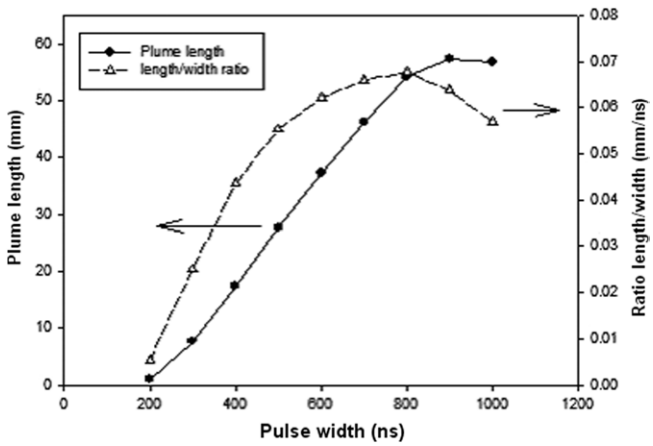

Figure 4. Plume length and ratio length/pulse width as a function of the pulse width of the applied voltage. Helium flow rate = 7.7 L min^{-1}, voltage = 5 kV.

the length for different gas flow rates. The measurements were carried in our laboratory (around sea level) where a relative humidity of 40% and a room temperature of 23 °C are typical. The length increases with the voltage, but eventually levels out at some point. A mechanism involving chemical reaction rates and diffusion rates is probably the cause of this plateau that leads to an equilibrium between the energy put into the plasma by the pulses and losses by diffusion and recombination.

We can see also from the curves that an optimal value for the gas flow exists: at 7.77 L min^{-1}, the plume reaches a maximum length. To confirm the existence of an optimum value for the gas flow, the plume length was plotted as a function of the gas flow rate for different voltages (see figure 2). As expected, the maximum value is obtained with a flow rate of about 7 L min^{-1}. The influence of the voltage is not important for very low or very high flow rates. However, to obtain a

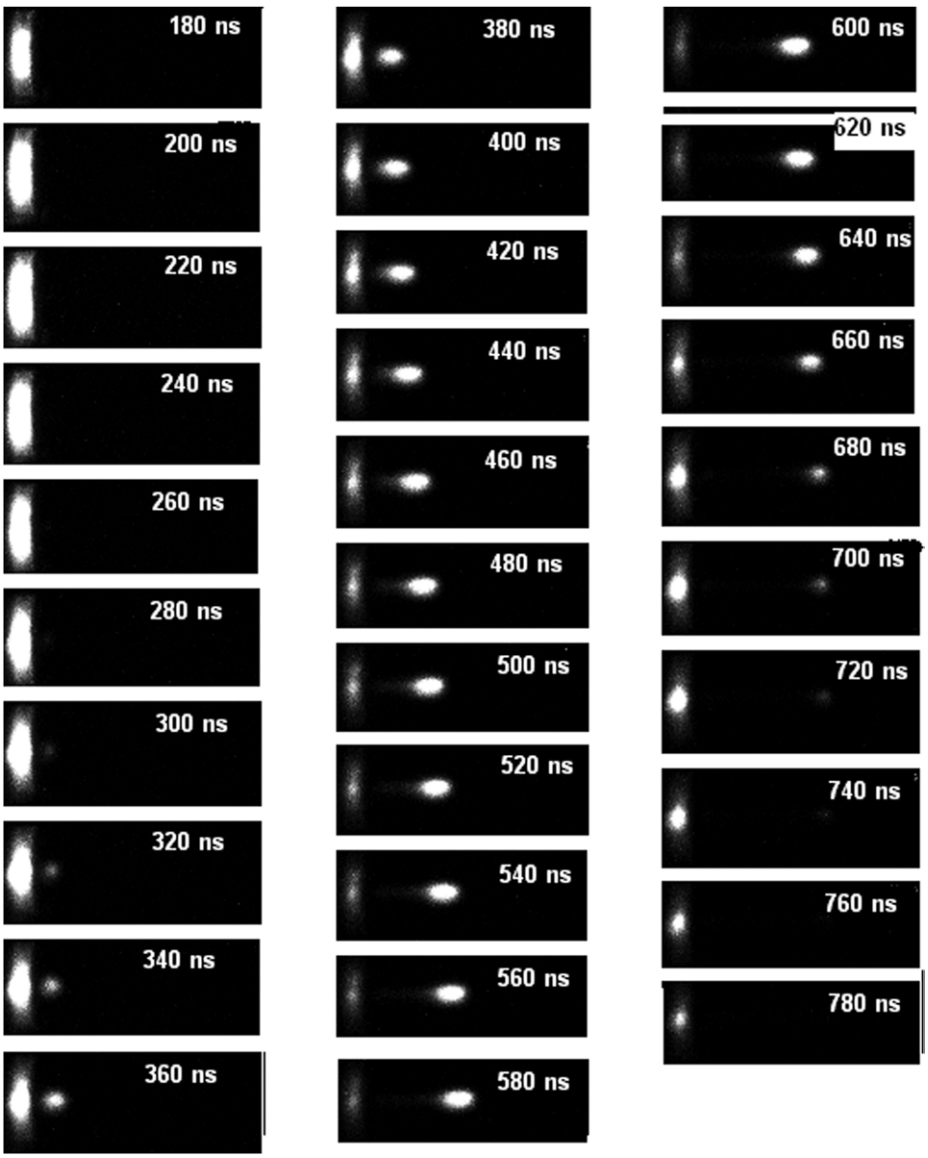

Figure 5. High-speed photographs of the discharge and the bullet emitted by the plasma pencil. Exposure time of the camera is 50 ns. $V = 5$ kV, pulse width is 500 ns.

greater plume length for increasing voltages, the gas flow rate value has to be close to 7 L min^{-1}. At higher flow rate values, the plume becomes unstable, then begins to decrease due to flow turbulence, as shown in figure 3.

Another very important parameter influencing the plume length is the pulse width of the applied voltage. An increase in the pulse width results in an increase in the length of the plume as seen in figure 4. The maximum length is reached for a pulse width of 900 ns, then the length decreases slightly with longer pulses. Between 200 and 800 ns, the increase in the plume length is quasi-linear which means that the length and the energy deposited into the plume are directly related. The ratio between the length of the plume and the pulse width indicates that the best efficiency is reached for a pulse width of 800 ns. Afterwards the increase is limited by the chemistry of the discharge in the air.

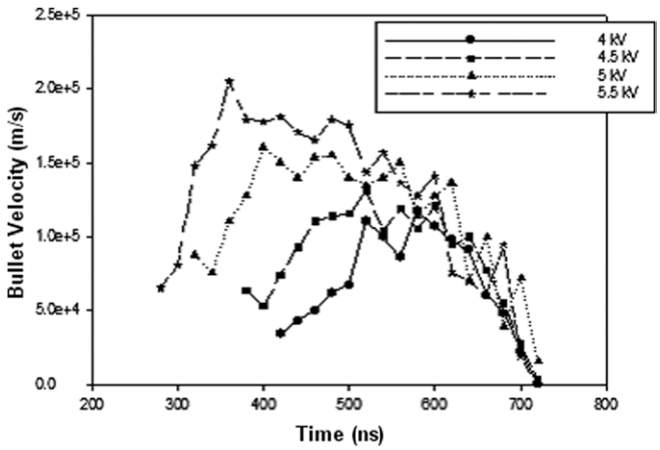

Figure 6. Bullet velocity versus time for different voltages, helium flow rate around 7 L min^{-1}. Pulse width is 500 ns.

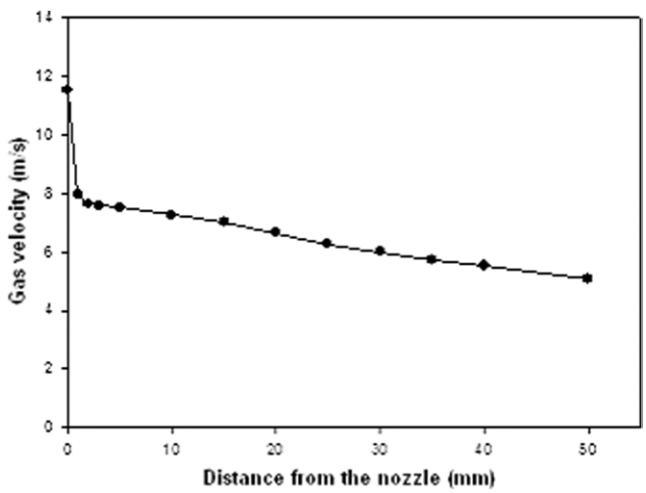

Figure 7. Gas velocity along the axis for a flow rate of 7.7 L min^{-1}.

3. Bullet formation, structure and propagation

The ignition and propagation of the bullets were investigated using a DiCAM-PRO ICCD camera. Figure 5 shows pictures of the discharge inside the discharge chamber (between the electrodes) and that of the bullet captured at different times.

As seen in figure 5, a first discharge is observed in the discharge chamber. This discharge occurs around the time the voltage pulse reaches its plateau (the rise time is about 50 ns). This discharge is accompanied by a first current peak. 120 ns after the occurrence of this peak a bullet is launched out of the chamber and into the ambient air. The launch of the bullet comes with a second current pulse that is much weaker than the previous one. When the voltage falls, a second discharge is ignited inside the chamber and is accompanied by a negative current pulse. It is at this time that the bullet extinguishes. Therefore, the propagation of the plume is only sustained during the voltage pulse. This was confirmed by changing the pulse width and finding that the bullet always extinguishes at the end of the applied voltage pulse. Another interesting observation is the fact that regardless of the magnitude of the applied voltage pulse, the bullet extinguishes at the same time, which also corresponds to the fall of the pulse. Figure 6 illustrates this observation. Figure 6 also shows that when the voltage increases, the average bullet velocity also increases. The shape of the curve remains the same, but the maximum velocity is reached earlier for higher voltages. Most interestingly, as the voltage increases the bullet formation occurs at earlier times with the extinguishing time remaining the same, as mentioned earlier. Therefore, in order to reach the end of its trajectory always at the same time, both the average velocity of the bullet and its formation time appear to change accordingly: earlier formation time and reaching higher velocities for increasing voltages. It is important to mention here that the velocity of the bullet is several order of magnitudes higher than the gas velocity out of the device. Figure 7 shows the gas velocity (flow rate of 7.7 L min^{-1}) as a function of the distance as measured by a Pitot tube. Figure 7 also shows that

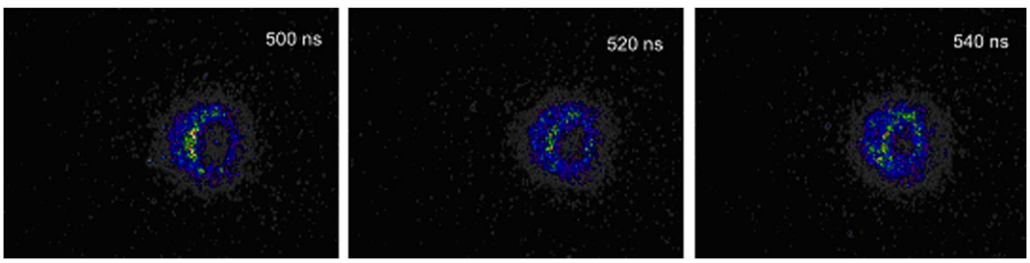

Figure 8. Photographs of the bullets illustrating their donut shape.

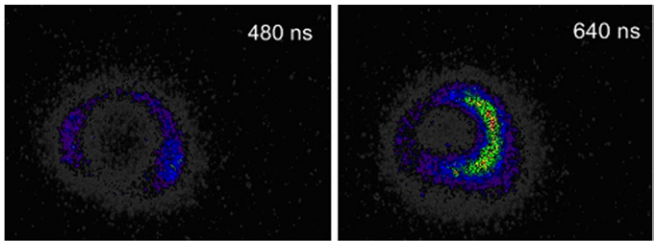

Figure 9. On-axis photographs of the plume at two different times, illustrating a donut shape.

the spatial dependence of the gas velocity is quite different from that of the plasma bullet.

To reveal the accurate features of the structure of the bullets, the ICCD camera was placed both at an angle and also head-on with respect to the plasma plume. It turned out that the bullet is, in fact, hollow and has a 'donut' structure. This is a very important observation as it supports the idea that the plasma bullet, in fact, propagates along the interface of two media, the helium channel and the ambient air. This leads to the proposal that the bullet is a solitary ionization surface wave. These types of waves were, in fact, predicted in low temperature plasmas by early investigators [5]. Figures 8 and 9 illustrate the donut shape of the bullets.

If the bullets are solitary surface waves then their velocity of propagation depends on the dielectric constant of the two adjacent media. The higher the dielectric constant of the outer layer, the lower the speed. In order to verify this we carried out the following experiment. Using one of our jet devices we attached a short glass tube to the exit of the device. This way the plume would propagate first through the tube before exiting to the ambient air. In this configuration, the bullets travel first in a helium–glass interface, then in a helium–air interface. Figure 10 shows the behaviour of the bullet velocity. Indeed, the bullet velocity increased as soon as the bullet exited the tube and entered the ambient air environment. This observation appears to support the proposal that the plasma bullets are, in fact, formed by a surface wave propagating at the interface between two media.

4. Control of the plasma bullets by an external dc electric field

The application of an external dc electric field affects the dynamics of the plasma bullets. Figure 11 shows a plot of the bullet velocity profile for various amplitudes of an externally applied dc voltage (applied between two plates parallel to the plume: E-field is orthogonal to the plume axis). It is seen that as the dc voltage increases the average velocity decreases and the time of first detection of the bullet increases. In addition, it was observed that the total length of the plume decreases. This was observed independently of the direction of the electric field (three directions were tried: along the plume axis, at 90° and at 180°), although the highest relative decrease in length was observed in the 180° direction (when the positive of the high dc voltage is near the outer electrode of the device, which is grounded). An interesting outcome of this experiment is the fact that if the external electric field is applied in a perpendicular direction to the axis of the plume (at 90°), it is observed that the plume gets deflected in the opposite direction of the field (towards the positive voltage). Our interpretation of this is that the overall charge of the plasma plume is negative, with the negative charges coming from electrons and negatively charged ions such as O_2^-. This is illustrated in figure 12.

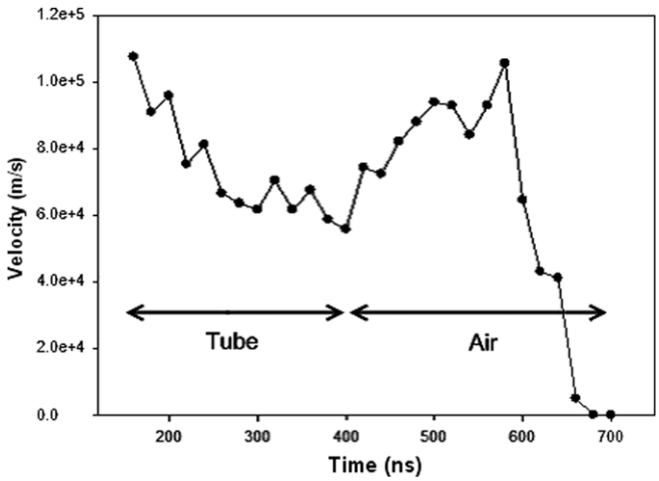

Figure 10. Bullet velocity with propagation in a tube and in the air, helium flow rate = 7.7 L min^{-1}, voltage = 5.7 kV. The pulse width is 500 ns.

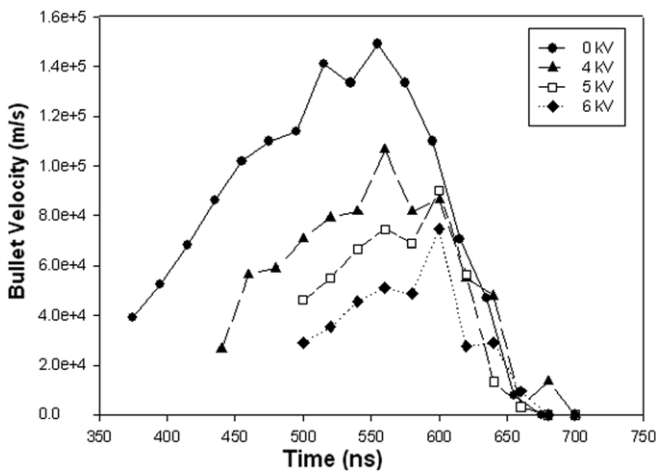

Figure 11. Bullet velocity profile for various externally applied dc voltages. The dc voltages are applied to establish a dc electric field perpendicular to the axis of the plume. Pulse width is 500 ns.

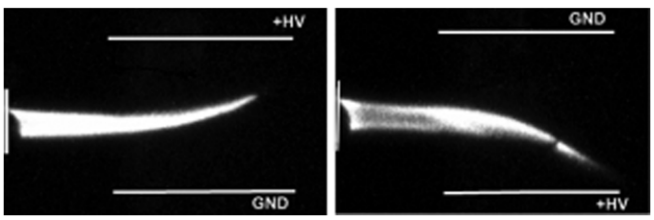

Figure 12. Images showing the plume bending towards the positive side of an externally applied dc voltage.

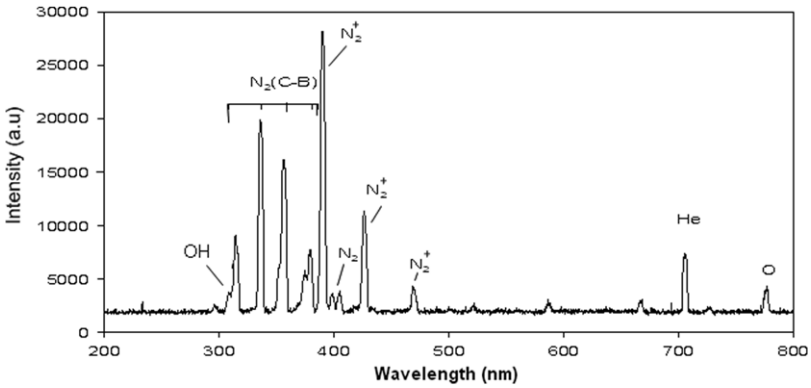

Figure 13. A typical emission spectrum from a helium plume emitted in ambient air. $V = 5$ kV and pulse width is 500 ns.

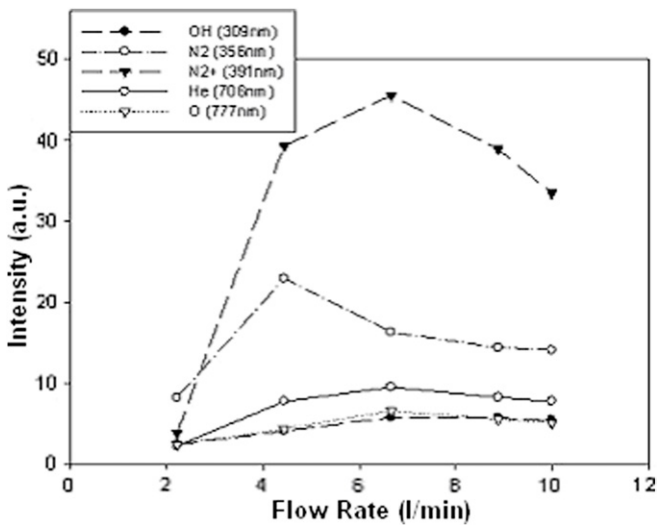

Figure 14. Intensities of selected emission lines as a function of the gas flow rate. Light is collected at 8 mm from the beginning side of the plume. $V = 5$ kV and pulse width is 500 ns.

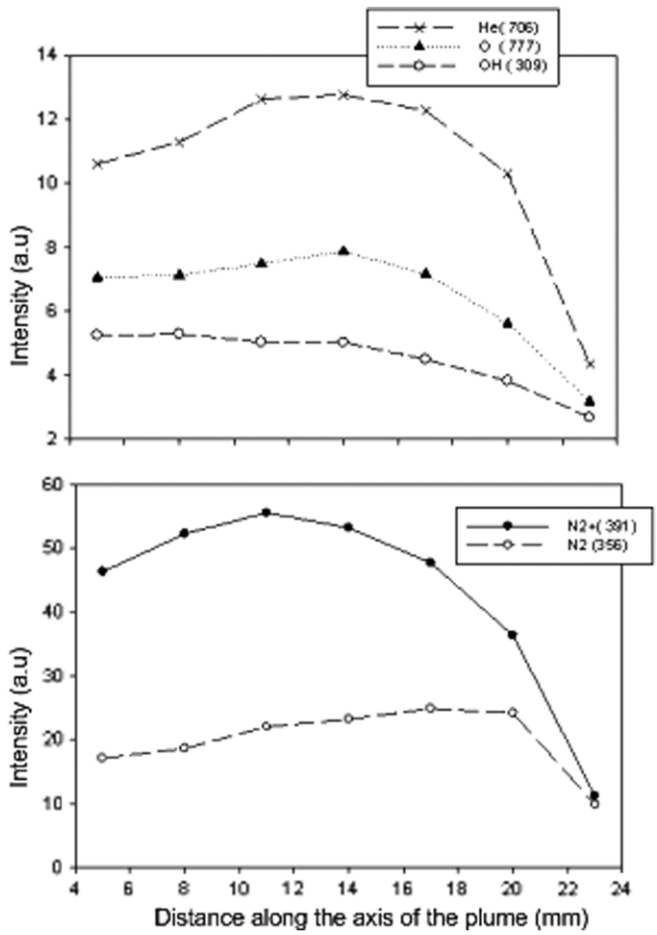

Figure 15. Intensities of the He, N_2, N_2^+, O and OH emission lines as a function of the axial distance from the beginning side of the plasma plume. Gas flow rate is 7 L min^{-1}. $V = 5$ kV and pulse width is 500 ns.

5. Role of the plume chemistry in its propagation

Optical emission spectroscopy was used to identify the chemical species present in the plasma plume. Figure 13 shows a typical emission spectrum for a helium plasma plume in air, taken by an Ocean Optics mini-spectrometer USB4000. The emission spectrum is dominated by N_2^+ and also by N_2 excited states. Atomic oxygen, O and OH lines are also present. To study the variation of the intensity of the main emission lines (N_2, N_2^+, O, OH and He) we used an optical fibre to collect the localized emission along the axis of the plume. Figure 14 shows the intensities of these emission lines as a function of the flow rate (collected at an axial distance of 8 mm from the outer aperture of the device) and figure 15 their intensities as a function of the distance along the plume axis (gas flow rate of about 7 L min^{-1}). The intensity curve of N_2^+ is particularly interesting: the N_2^+ line intensity reaches a maximum value at the same flow rate ($\sim$7 L min^{-1}) under which a maximum plume length is achieved. This is an indication that the N_2^+ density is a crucial parameter influencing the physical characteristics of the plume. Of note is also the fact that the helium line follows the same behaviour with a maximum at the 7 L min^{-1} flow rate. Figure 15 shows that the N_2^+ and He line intensities exhibit optima around 12 mm from the beginning of the plume. This position happens to be around the same position where the bullets reach their maximum velocity. This fact seems to indicate that N_2 ionization by helium metastable states may play an important role in the propagation mechanism of the plume/bullets.

6. Conclusions

Using fast photography and spatially resolved emission spectroscopy, insights into the nature and dynamics of the plasma bullets emitted by a cold plasma jet have been presented. The spatial distributions of several chemical species of interest and their dependence on the flow rate have been highlighted. Fast photography revealed that the time of formation of the bullets and their speed are strongly dependent on the magnitude of the applied voltage, that the time at which the bullets stop propagating is set by the fall of the voltage pulse and, most importantly, that the bullets exhibit a hollow, donut-shaped structure. This indicates that ionizing surface waves may be the physical process behind the creation and propagation of the bullets. A new and interesting feature was also presented: the control of the initiation time and length of travel of the plasma bullets by an externally applied dc electric field.

Acknowledgment

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