Pure air-plasma bullets propagating inside microcapillaries and in ambient air

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Abstract.

This paper reports on the characterization of air-plasma bullets in microcapillary tubes and in ambient air, obtained without the use of inert or noble gases. The bullets were produced by nanosecond repetitively pulsed (NRP) discharges, applied in a dielectric barrier discharge (DBD) configuration. The anode was a tungsten wire with a diameter of $50\text{ }\mu\text{m}$, centered in the microcapillary, while the cathode was a silver ring, fixed on the outer surface of the fused silica tube. The effect of applied voltage and inner diameter of the microcapillary tube on the plasma behavior were investigated. Inside the tubes, while the topology of the bullets seems to be strongly dependent on the diameter, their velocity is only function of the applied-voltage amplitude. In ambient air, the propagation of air-bullets with a velocity of about $1.25 \times 10^5\text{ ms}^{-1}$ is observed.

1. Introduction

Guided streamers, so-called plasma bullets, in ambient air, have been studied extensively in the last decade [1–5]. The main application fields of these non-equilibrium plasmas propagating out of the interelectrode region are bio-medicine [6–8] and surface treatment [9–11]. These applications are temperature sensitive, therefore, the control of gas heating by the plasma generation is a key point. A convenient way to create a non-thermal plasma at atmospheric pressure is the use of noble or inert gases, in order to reduce the breakdown voltage, in a dielectric barrier discharge (DBD) configuration, which restricts the conductive current. Thus, in the large majority of previous studies, the discharges were generated in pure or mixture-containing helium [1–5], argon [12, 13], neon [14, 15], or krypton [16]. However, the high cost of these gases could be an issue for some applications.

On the other hand, in air or in air-containing mixtures, non-thermal plasma discharges at atmospheric pressure have also been extensively studied for numerous applications such as flue gas treatment [17], plasma-assisted combustion [18] or biomedical applications [19]. However, to our best knowledge, the generation of pure air-plasma bullets by non-thermal plasma discharges at atmospheric pressure has never been reported. This is the purpose of the present study.

2. Experimental setup

The air-plasma bullets were produced by nanosecond repetitively pulsed (NRP) discharges in a DBD configuration. Figure 1 presents the two experimental configurations used in this study. The anode is a tungsten wire of 50- μm diameter, centered in the microcapillary, while the cathode is a silver ring, fixed on the outer surface of the tube. In order to avoid parasitic discharges outside the microcapillary, a 1-mm thick insulation layer covers the silver ring. The tubes used are out of fused silica (IDEX Corporation). They have inner diameters (IDs) of 100, 150, 300 and 500 μm , *i.e.* in the same range as the typical diameter of discharges in air at atmospheric pressure [20, 21]. The corresponding thickness of the tubes are comprised between 115 and 200 μm . The microcapillary is fed with a 4-sccm flow of air at atmospheric pressure, inducing a flow velocity always slower than 10 ms^{-1} . The injected gas is dried air, obtained by compression and refrigeration process (down to 233 K). The gap distance is kept constant at $1.35 \pm 0.15\text{ mm}$. For studying the bullets propagation inside the tube, the length of the microcapillary after the cathode is fixed at 5 cm [see figure 1(a)]. For the generation of the plasma bullets in ambient air [see figure 1(b)], the length of the tube after the silver ring is about 1 mm. In order to avoid propagation of streamers on the outer surface of the microcapillary, a square plate of teflon ($2 \times 2\text{ cm}^2$, thickness 0.8 mm) is fixed perpendicularly to the tube exit.

The high-voltage pulses of 10-ns duration and 5.2–8.2 kV amplitude, at a repetition frequency of 1 kHz, were generated by a solid-state pulse generator (FPG 10-30NM10, FID GmbH). Voltage and current across the electrodes were measured with appropriate probes (a Tektronix 75 MHz high-voltage probe, model P6015A, and a Pearson coil, model 2877). Both signals were recorded simultaneously with a 500 MHz bandwidth oscilloscope (Agilent Technology DSOX3054A). The energy deposition per pulse was determined from these electrical measurements, following the method detailed by Pai [22] and summarized elsewhere [23]. First, a synchronization between the current and voltage signals was performed. Indeed, any cable or probe affects differently the lag time between the discharge events and measurements. For nanosecond duration pulses, this lag time is significant and a synchronization is necessary. For this purpose, in this study, the current waveforms were shifted by 6.5 ns. Then, the current and voltage signals were multiplied and integrated to obtain the energy deposition per pulse. An example of synchronized voltage and current waveforms, and the corresponding energy per pulse, is presented in figure 2. The beginning of the high-voltage pulse defines the

reference time, t_0 . The ringing in both the voltage and current signals is due to parasitic impedance and capacitance in the electric circuit. The subsequent resulting error on the energy determination is estimated to be of about 20 %. Therefore, the energy per pulse should be taken only as an indicator, not allowing further analysis.

The propagation velocity of the bullets was determined with the aid of an intensified charge coupled device (ICCD) camera (Princeton Instruments PIMax, 1024×1024 pixels²), equipped with a zoom lens (Thorlabs, Navitar 1-50486). Each image was obtained by accumulating over 20,000 events. The exposure time was 2 ns. The images were synchronized with the discharges using a function generator (Tektronix AFG 3021). The jitter between two pulses was very low, less than 0.5 ns, and the synchronisation accuracy between the camera and the plasma generator was about 0.2 ns. Taking into account the propagation speed of the observed phenomenon, the scattering effect due to accumulation and synchronization can be neglected. For a given image, the position of the discharge front was defined as the right part of the bullet where the pixel intensities were about 20 % of the maximum intensity of this particular image. In order to facilitate visualization of the plasma, the greyscale images have been mapped to a color lookup table.

The gas temperature of the plasma discharge has been investigated by optical emission spectroscopy (OES) analysis. The optical setup comprised an Acton SpectrPro-320i spectrometer (focal length 320 mm, grating of 1200 gr/mm, blazed at 300 nm) fitted with a 1024×1024 pixels² ICCD camera (Princeton Instrument PIMax) and an imaging fiber adapter (Acton FC-446-010) combined with an optical fiber (Acton LG-4550-020). The full width at half maximum (FWHM) of the slit function was 0.83 nm. All the light from the plasma zone, including the interelectrode area, was collected. Each spectrum was obtained by accumulating over 100,000 events. The exposure time was 2 ns, synchronized with the plasma discharges, by using a function generator (Tektronix AFG 3021).

3. Results and discussion

Figures 3(a)–(f) present the discharge dynamics out of the inter electrode region for a 100- μ m ID microcapillary. The applied voltage was 7.2 kV and the energy per pulse 85 μ J. The fast-moving discharges obtained have a luminous front filling the whole section of the microcapillary. They propagate from the cathode area to the outlet of the tube, in the direction of the air flow. This behavior, similar to plasma bullets in helium, allows us to call these discharge fronts, air-plasma bullets. Figure 3(g) shows air-plasma bullets obtained with a 500- μ m ID microcapillary. In this case, the discharges propagate at the vicinity of the surface. This effect of the microcapillary diameter on the topology has already been observed by Jansky *et al.* [24], for similar discharges propagating between electrodes. They explained that, in air, due to the relatively high permittivity of the capillary material, the electric field increases close to the surfaces. Then, the discharges naturally propagate close to the surfaces. However, when the

diameter of the microcapillary is of the order of the typical size of the discharge, it is constricted by the surfaces and appears homogeneous. These effects could also explain our present results.

The duration of light emission from the plasma region is about 25 ns. Knowing the delay between two images, the bullets velocity can be determined. Figure 4 presents the influence of the microcapillary ID on the temporal evolution of the bullet velocity. The reproducibility of the experiments has been verified by repeating three times each measurement. The error bars represent the uncertainty in estimating the front of the discharges from the ICCD images. These results have been obtained for an applied voltage of 8.2 kV and an energy per pulse of 115 μ J. At 1 kHz, the corresponding average power is then 115 mW. First, even if the shape of the bullets depends on the microcapillary ID, this parameter does not seem to affect their velocity. Indeed, regarding the measurement uncertainty, for IDs from 100 to 500 μ m, between 8 and 14 ns after t_0 , the velocity of the bullets is about $4 \times 10^5 \text{ ms}^{-1}$. Then, it decreases rapidly to less than $1 \times 10^5 \text{ ms}^{-1}$, and finally the bullets vanish. This range of velocities corresponds to the one obtained by Jansky *et al.* [24] for similar conditions, between electrodes.

The effect of the applied voltage has been investigated for a 100- μ m ID microcapillary. Figure 5 shows that the maximum velocity of the bullets strongly depends on the applied-voltage amplitude. For a delay of 10 ns after the beginning of the pulses, the velocity of the air-plasma bullets is four times higher at 8.2 kV than at 5.2 kV (5.2×10^5 and $1.2 \times 10^5 \text{ ms}^{-1}$, respectively). This behavior is in agreement with previous results obtained in helium [25]. A 5-mm propagation length of the air-plasma bullets inside the microcapillary has been obtained for an applied voltage of 8.2 kV. This length could probably be increased by applying longer high-voltage pulses (typically a few hundreds of nanoseconds, as in references [25, 26]). Finally, the gas temperature has been determined from spectroscopic analysis of the N_2 (C-B) transition (obtained with a methodology detailed elsewhere [23]). Figure 6 presents an example of experimental spectrum, obtained for a 100- μ m ID microcapillary and an applied voltage of 8.2 kV, at the plasma-emission peak, and associated simulations performed with the SPECAIR software [27, 28] for a vibrational temperature of 3500 K, and rotational temperatures, T_{rot} , of 300, 400 and 500 K. Thus, for this experimental condition, the heating of the gas by the discharges has been found to be about 100 K. During the 25 ns of the N_2 (C-B) emission, all the recorded spectra were similar, with no significant temperature evolution. Thus, the thermal impact of the discharges seems to be reasonable. However, further investigation would be necessary to conclude about the temperature of these air-plasma bullets.

Hofmann *et al.* [29] have shown the propagation of pure argon-plasma bullets in an argon atmosphere, at atmospheric pressure. They explained this result by a memory effect due to previous pulses. Indeed, by adjusting the frequency of the pulses and the flow velocity, guiding of subsequent streamers is possible, leading to the propagation of plasma bullets. In pure helium, due to a combined low electronic density in the head of

the streamer and slow propagation of the ionization front [30], they did not observe the propagation of bullets, but a diffuse and homogeneous plasma.

In order to determine the plasma behavior in pure air, a similar setup [see figure 1(b)], with a 150- μm ID microcapillary opening on a dielectric plane, was used. Figure 7 presents images of the observed discharge behavior. Air-plasma bullets clearly propagate in ambient air, from the outlet of the capillary, in the direction of the main flow. Considering the low speed of this flow (less than 10 ms^{-1}), this behavior cannot be explained as in the work of Xian *et al.* [31] by a fast convection of the plasma generated between the electrodes. In air, the memory effect could also be responsible for the guidance of the streamers. Indeed, the pre-ionization level of the flow (mainly O_2^- and O_4^+ at room temperature [32, 33]), induced by the previous pulses, could be high enough to generate a confined plasma propagating in the air-jet. Tholin *et al.* [34] have shown that for similar NRP discharges in ambient air, taking into account a detailed chemistry of charged species during the inter-pulse time, the ion density, 1 ms after a pulse, remains high, about 10^{10} cm^{-3} .

In parallel, a surface discharge develops on the dielectric plate, in a quasi-perfect circular shape. For 7.2-kV applied voltage, a circle of about 6-mm diameter is formed in 10 ns, which correspond to an average propagation speed of $1.4 \times 10^6\text{ ms}^{-1}$. One reason for the development of this surface discharge could be a local increase in the electric field close to the teflon surface, but to explain this effect, further investigations are required.

Figure 8 presents the influence of the applied voltage on the air-plasma bullets velocity in ambient air. Unlike the plasma confined in microcapillaries, in the range of 6.2 to 8.2 kV, the bullets velocity is not significantly affected by the applied voltage, with an average propagation velocity of $1.25 \times 10^5\text{ ms}^{-1}$. Typically, a 4-mm propagation length of the air-plasma bullets in ambient air has been obtained for 7.2-kV applied voltage.

4. Conclusion

This study puts forward the generation of air-plasma bullets at atmospheric pressure inside dielectric microapillaries and in ambient air, without the use of inert or noble gases. For inner diameters of the dielectric tubes in the range of the typical diameter of the discharges, actually between 100 and 500 μm , electrically-driven streamer propagation outside the electrode gap has been observed. It has also been shown that, even if the shape of the bullets depends on the microcapillary inner diameter, this parameter does not significantly affect their velocity. Conversely, the plasma bullets velocity depends on the applied voltage inside the tubes but not much in ambient air.

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Figure 1. (a) Setup for air-plasma bullets in microcapillaries, with electrical diagnostics and ICCD camera, (b) setup for air-plasma bullets in ambient air.

Figure 2. Example of voltage and total current measured for NRP discharges applied at 1 kHz, gap distance 1.4 mm, 150- μm ID capillary, and the corresponding energy per pulse.

Figure 3. Visualizations of the propagation of air-plasma bullets inside a 100- μm ID microcapillary (a–f), and inside a 500- μm ID microcapillary (g), for 7.2-kV applied voltage.

Figure 4. Influence of the inner diameter (ID) of the microcapillary on the temporal evolution of the air-plasma bullet velocity, for 8.2-kV applied voltage.

Figure 5. Influence of the applied voltage on the temporal evolution of the air-plasma bullet velocity in a 100- μm ID microcapillary.

Figure 6. Example of experimental spectrum of the $\text{N}_2(\text{C-B})$ transition and simulations performed with the SPECAIR software [27], for an ID of 100 μm and 8.2-kV applied voltage.

Figure 7. Visualizations of the air-plasma bullets propagating in ambient air, obtained with a 150- μ m ID microcapillary and 7.2-kV applied voltage.

Figure 8. Influence of the applied voltage on the temporal evolution of the air-plasma bullet velocity in ambient air (150- μ m ID microcapillary).