Eur. Phys. J. Appl. Phys. (2016) 75: 24713
DOI: 10.1051/epjap/2016150594

THE EUROPEAN PHYSICAL JOURNAL APPLIED PHYSICS

Regular Article

# Impact of an atmospheric argon plasma jet on a dielectric surface and desorption of organic molecules*

Xavier Damany, Stéphane Pasquiers[a], Nicole Blin-Simiand, Gérard Bauville, Blandine Bournonville, Michel Fleury, Pascal Jeanney, and João Santos Sousa

LPGP, CNRS, Univ. Paris-Sud, Univ. Paris-Saclay, 91405 Orsay Cedex, France

Received: 1 December 2015 / Received in final form: 9 March 2016 / Accepted: 26 April 2016


**Abstract.** The propagation of a DC-pulsed argon plasma jet through the surrounding ambient air, and its interaction with an ungrounded glass plate placed on the jet trajectory, was studied by means of fast imaging. The surface plays an important role in the spatio-temporal characteristics of the plasma. Indeed, for an argon jet propagating perpendicularly to the surface, the plasma jet structure changes from filamentary to diffuse when the distance between the nozzle of the capillary tube and the surface is short ($\leq$10 mm). Changing the angle between the capillary tube and the glass plate, and varying the gas flow rate strongly affects the spatial extension of the plasma that develops on the surface. This surface plasma propagates while the plasma in the argon jet is maintained with the same luminous intensity. Finally, this plasma jet shows interesting characteristics for desorption of low volatile organic molecules such as bibenzyl. A maximum removal of bibenzyl is located at the intersection area between the jet axis and the glass surface, and some of the initially deposited molecules are found intact in gas phase.

## 1 Introduction

As emphasised by recently published review articles [1–3], low temperature atmospheric plasma jets using rare gases (He, Ar) have been the subject of numerous studies for several years, due in particular to their interest for different types of applications like material processing or biomedicine [4–11]. These micro-plasmas can also be used in chemical analysis with mass spectrometry, for example in the detection of low volatile organic molecules adsorbed on surfaces [12–14]. For such an application, the understanding of the interaction between the plasma jet and the surface is of great importance. In the present work, an argon jet propagates in ambient air and meets an ungrounded glass plate. A HV-pulsed dielectric barrier micro-discharge is used to generate guided streamers propagating in the rare gas and impacting the solid surface. The effect of this obstacle on the spatio-temporal characteristics of the plasma is described using a fast imaging technique capturing the total plasma emission. The role of three important experimental parameters is studied in more detail: (i) the distance between the nozzle of the capillary tube and the surface, (ii) the angle between the axis of the capillary tube and the surface, (iii) the argon gas flow rate. The effect of the plasma jet on a sample of bibenzyl ($C_{14}H_{14}$) deposited on a glass plate is examined using a binocular magnifying-glass and gas chromatography (GC-MS).

## 2 Experimental set-up and electrical parameters

The plasma jet is generated from a dielectric barrier discharge (DBD) like those previously used in references [15,16]. It is made of a quartz capillary tube with inner and outer diameters of 1.7 mm and 4.3 mm, respectively. A grounded copper foil electrode, 1 cm long and 100 $\mu$m thick, is wrapped around the external side of the dielectric at a distance of 5 mm from the nozzle. A capillary electrode, with an inner diameter of 1.2 mm, is glued inside the dielectric tube at a distance of 10 mm from the nozzle.

The DBD is driven by high-voltage pulses produced by a homemade power supply, and the discharge electrical parameters are measured using adapted probes connected to a digital oscilloscope. The spatio-temporal evolution of the plasma emission is investigated using an ICCD camera (Princeton Instrument PIMAX-3) equipped with a UV-macro lens (EADS Sodern CERCO2178 F/2.8,

[a] e-mail: stephane.pasquiers@u-psud.fr

* Contribution to the topical issue "6th Central European Symposium on Plasma Chemistry (CESPC-6)", edited by Nicolas Gherardi, Ester Marotta and Cristina Paradisi

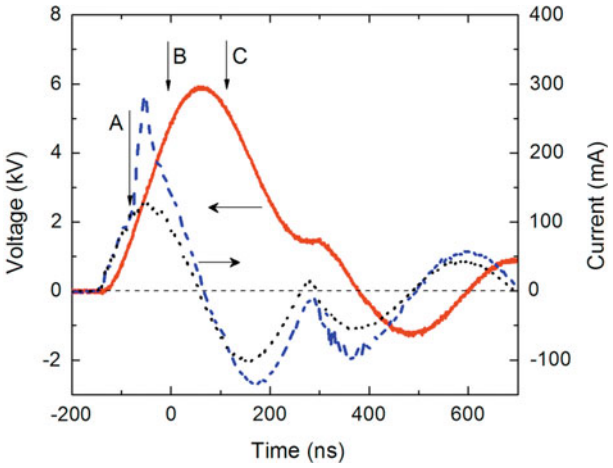


**Fig. 1.** Time evolution of the current (total current: blue dashed line, displacement current: black dotted line) and the voltage (red solid line) for an applied HV pulse of 6.0 kV at 20 kHz. Arrows A, B, C, please refer to the text.

spectral range 220–900 nm). The optical axis of the camera is placed perpendicular to the axis of the capillary tube.

An example of an electrical recording is given in Figure 1, for a peak value of the applied voltage to the DBD of 6.0 kV at a repetition rate of 20 kHz, and for an argon flow rate of 0.7 L/min NTP. For the electrical parameters used in the measurements discussed in the present article (6 kV, 20 kHz), the HV pulse duration is 215 ns (FWHM) and the electrical energy deposited in the discharge is about 20 $\mu$J/pulse. The moment of the discharge breakdown is marked by the arrow "A" in Figure 1. The measured current before this instant is due to the circuit capacitance (2.3 pF). The displacement current (without discharge) is represented by the black dotted line.

The desorption of molecules by plasma jet may concern the detection of prohibited substances such as narcotics or explosives [17]. Many of these substances contain aromatic rings. We have studied the effect of our plasma jet on the desorption of 1,2-diphenylethane also called bibenzyl, $(C_6H_5\text{-}CH_2)_2$, because this aromatic chemical compound can be regarded as a model molecule due to the presence of two phenyl groups. Bibenzyl is dissolved in methanol in order to obtain solutions with known concentrations. A precise number of drops of one of these solutions are deposited on glass plates by means of a syringe controlled by a pump, allowing reproducible deposits. Methanol is then evaporated and a deposit of a solid bibenzyl is obtained in a controlled amount. The distribution of the deposit on the surface is more homogeneous when a drop of a highly concentrated solution is used. Thus, we chose to work with one drop of a solution of bibenzyl at 5000 ppm in methanol, producing a 250 $\mu$g of solid deposit of bibenzyl.

The visible effect of the plasma on the deposit is measured using a binocular magnifying-glass. Additionally, samples are extracted from the gas phase during plasma treatment using Tenax® tubes containing a resin to absorb and concentrate the molecules. One end of the tube is directed towards the deposit with an angle of 45° whereas the other is connected to a pump. This system allows trapping molecules desorbed by the plasma during the whole treatment. These samples are then injected by means of a thermo desorber (Alyxan) in a chromatograph and analyzed by GC MS (Varian Saturn 2000 – column WCOT ulti-metal 25 m × 0.25 mm, coating CP SIL PAH – CB).

In Table 1 are given the range of values used for the four experimental parameters considered in this work.

The individual role of each parameter has been studied keeping all others constant. The data discussed in the following are representative examples of the experimental results that have been obtained.

## 3 Free propagation of the argon jet (no obstacle)

In Figure 1, the arrow "B" indicates the moment when a streamer begins to propagate outside the capillary tube. With no obstacle on the path of the argon jet, streamers are able to propagate up to a maximum distance of 30 mm from the nozzle for a gas flow rate of 0.7 L/min NTP, as it can be seen in Figure 2, in which four ICCD camera recordings for an exposure time of 200 ns are displayed. Each image corresponds to a single HV pulse, and the measurement of the plasma emission captured the whole propagation length of the streamers. The quartz tube is located at the top of images (marked in white). Whatever the parameters values (gas flow rate, applied voltage, pulse repetition frequency), the plasma outside the tube appears filamentary with some branching phenomena.

Figure 3 shows a camera recording at the instant marked by the arrow "C" in Figure 1, taken with an exposure time of 20 ns but accumulated over 10 successive HV pulses, giving details about how the plasma propagates in the thin channel of argon gas. A group of very intense streamer heads can be seen at 17.5 mm from the tube nozzle, while very small emission intensity is detected behind them, up to the capillary tube. The 10 streamers heads are distributed over a maximum distance of 4.5 mm.

**Table 1.** Range of the experimental parameters used in this work.

| | |
|---|---|
| Gas flow rate | 0.25 up to 1.0 L/mn NTP |
| Distance between nozzle and glass plate | 5 mm up to 15 mm |
| Angle between the tube axis and the glass plate | 25°, 45°, 90° |
| Duration of the interaction between the plasma and the bibenzyl deposit | 10 up to 60 s |

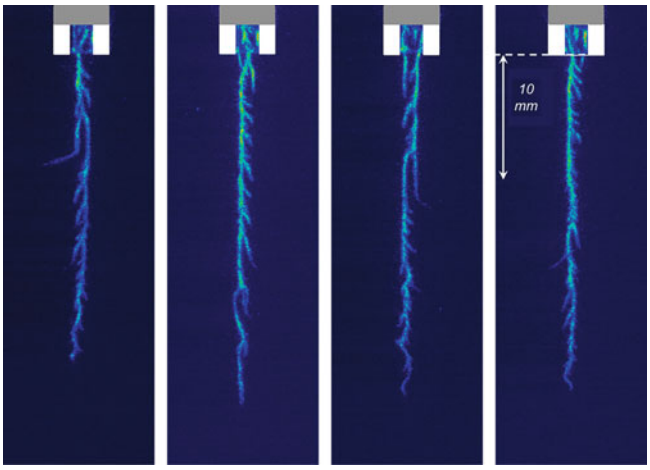


**Fig. 2.** Plasma emission captured by ICCD camera for an exposure time of 200 ns and for single HV pulses. Experimental parameters: 6 kV, 20 kHz, 0.7 L/min NTP, free argon jet.

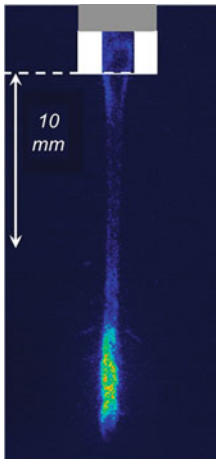


**Fig. 3.** Plasma emission captured by ICCD camera for an exposure time of 20 ns, and accumulated over 10 successive HV pulses. Same experimental parameters as those of Figure 2.

For the experimental conditions of Figures 1–3, a rough estimate of the propagation velocity of the streamers head was performed at four different instants, see Table 2.

The values of the velocity of the streamers head given in Table 2 corresponds to a mean instant velocity determined over 100 successive HV pulses, by considering the position of the maximum emission of the plasma (believed to be the mean position of 100 streamer heads, taken with an exposure time of 20 ns) between two time values separated by 40 ns for the first three velocities and 60 ns for the last. It appears that velocity of the streamers head slightly decreases during the propagation in the argon gas channel, up to 25 mm from the nozzle; for a higher distance it rapidly drops to zero at 30 mm. Such a phenomenon was also observed for other flow rates but no systematic precise measurement was performed during the present study. The data provided here are just indicative for 0.7 L/min NTP.

**Table 2.** Mean propagation velocity of the plasma in the argon jet for the experimental conditions of Figures 1–3.

| Time after discharge peak current (ns) | Mean distance of the streamers head from the nozzle (mm) | Estimated mean instant velocity (m/s) |
|---|---|---|
| 80 | 3 | – |
| 120 | 11 | $2.0 \times 10^5$ |
| 160 | 17.5 | $1.6 \times 10^5$ |
| 200 | 23 | $1.4 \times 10^5$ |
| 260 | 26.5 | $0.6 \times 10^5$ |

## 4 Physical effects of an ungrounded glass plate

### 4.1 Glass plate placed perpendicularly to the axis of propagation of the argon jet

When a glass plate (1 mm thick, ungrounded) is placed perpendicularly to the trajectory of the argon jet, the streamer impacts the surface and the plasma spreads over it. For a distance separating the plate from the nozzle, $d_{NP}$, longer than 10 mm, the discharge characteristics are similar to those measured for the free jet, including the occurrence of branching phenomena, except for the development of a plasma on the surface.

If $d_{NP}$ is shorter than 10 mm, with all other things being equal, the spatial distribution of the plasma appears more homogeneous in the small gaseous gap, as branching phenomenon disappears. This effect is illustrated in Figure 4, showing camera recordings for a $d_{NP}$ of 5 mm, at two different times: first, cases a1/(integrated over 10 HV pulses) and a2/(one HV pulse), when the streamer has just reached the surface, and second, case b/(10 pulses), 40 ns after cases a1/ and a2/.

For the case a1/ of Figure 4, it is found that a dark space is established just at the exit of the capillary tube over a distance of about 2 mm from the nozzle; a plasma emission is detected at its boundaries. However the emission appears more homogeneous at a closest distance to the plate, i.e., from 2.5 mm up to 5 mm. Although the detected emission intensity is lower in case a2/ compared to a1/, because the acquired image corresponds to a single HV-pulse (vs. 10 HV-pulses), it confirms the diffuse character of the plasma created in the argon gas when a glass plate is placed perpendicularly to the axis of propagation of the argon jet at $d_{NP}$ shorter than 10 mm.

The surface plasma appears circular to the eye. For the chosen experimental parameters, the diameter of this plasma reaches about five times the internal diameter of the capillary tube. From the measurements shown in Figure 4, a1/ and b/, the estimated propagation velocity of the plasma on the dielectric surface is of the order of 8 to $9 \times 10^4$ m/s, i.e., slightly lower than the streamers velocity in the free jet of argon gas (cf. Tab. 2).

ICCD measurements also reveal that the plasma emission is much more intense at the surface than in the gas volume separating the nozzle and the surface, though less homogeneous. A plasma emission is still measured well

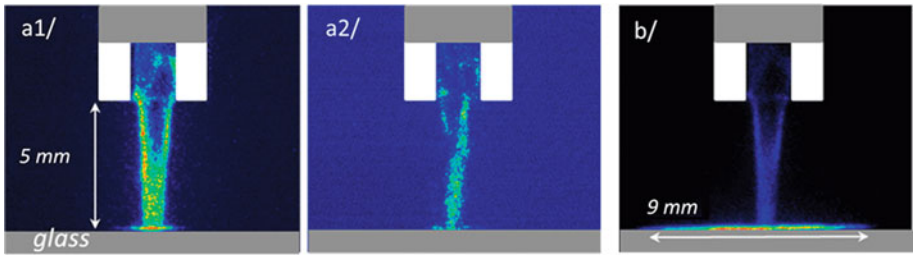


**Fig. 4.** Plasma emission captured by ICCD camera for an exposure time of 20 ns. Distance from nozzle to plate: 5 mm. Cases a1/, a2/, and b/: please refer to the text. Same experimental parameters as those of Figure 2.

after the discharge breakdown, both on the surface and in the gas volume. This emission should be related to the long lifetime of Ar electronic metastable states created by the plasma [18], these states being lost through quenching by molecules of the surrounding gases ($N_2$, $O_2$, $H_2O$).

### 4.2 Role of the angle between the axis of the argon jet and the glass plate

An important parameter in the present study is the angle between the argon jet axis and the glass plate. Examples of ICCD measurements are given in Figure 5, when the nozzle is placed at 5 mm from the plate, and the angle is 25°; this condition was chosen for the experiment illustrated by Figure 9 in Section 5.2. The gas flow rate was set to 0.25 L/min NTP (top image) or 1.0 L/min NTP (bottom image). For both images, the plasma emission was captured during 250 ns, i.e., during the full propagation time of the streamers, and accumulated over 10 HV pulses.

For the lowest gas flow rate value studied, 0.25 L/mn NTP, the plasma connects to the plate at different locations upstream of the intersection point between the axis of the capillary tube and the surface. The plasma develops at the surface below the main discharge propagating in the gas phase. However, when the gas flow rate is progressively increased up to 1.0 L/mn NTP, less and less streamers develop perpendicularly to the argon jet axis (there are less and less upstream plasma connections with the plate) and the surface plasma becomes mainly located downstream of the intersection point.

Time-resolved measurements of the plasma emission have been performed in order to describe more precisely the interaction of the plasma with the glass plate. Examples of results are shown in Figure 6, for a nozzle placed at 5.5 mm above the plate, for an angle of 45°, and for a gas flow rate of 0.25 L/min NTP (here the $d_{NP}$ value is slightly higher by only 0.5 mm than for Figure 5; it corresponds to the condition chosen for the experiment illustrated by Figure 8 in Section 5.2; note that the results obtained were at least qualitatively similar in the whole range of values of $d_{NP}$ studied, and for both angles 25° and 45°).

For the whole set of images, the plasma emission was captured during only 3 ns, but accumulated over 100 HV pulses. The time indicated on each image corresponds to

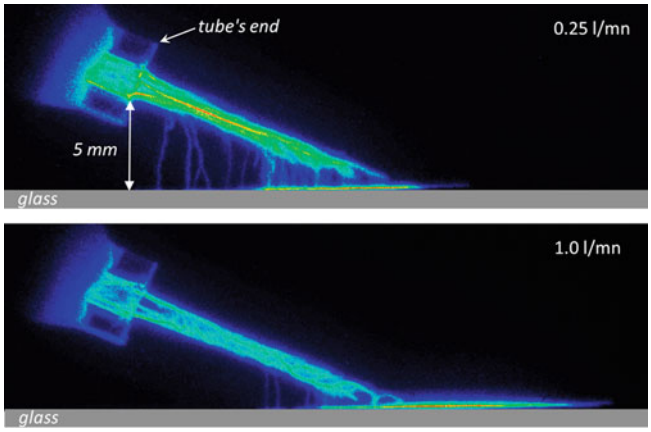


**Fig. 5.** Plasma emission captured by ICCD camera for an exposure time of 250 ns and accumulated over 10 successive HV pulses. Distance from nozzle to plate: 5 mm. Angle between the glass surface and the tube axis: 25°. Discharge parameters: 6 kV, 20 kHz. The argon flow rate values are given in the pictures.

the delay with respect to the instant at which the streamers begin to propagate outside the capillary tube (indicated by the arrow "B" in Fig. 1). The end of the tube is located in the upper left corner of the images. Intensities were normalized to the maximum value detected at each time, and are therefore not comparable between the different images. The first image, image A at 24 ns, shows that streamers initially propagate following the tube axis. The middle of the elongated reddish area corresponding to the maximum intensity (indicated by the arrow "a" in the image) is located at 2.7 mm from the nozzle; this area corresponds to the major location of the streamers heads for the selected 100 discharges. Six ns later, image B at 30 ns, the streamers are distributed over a wider area, with a symmetrical plasma emission in the argon jet along the tube axis. The maximum distance of propagation of the streamers from the end of the capillary tube is approximately 5.6 mm. Another 6 ns later, image C at 36 ns, streamers deviate towards the glass surface from their main propagation path, and some of them reach already the glass surface (indicated by arrow "b" in the image). After, image D at 42 ns, the plasma extends over the surface, in both directions from the main intercept point of the streamers with the glass; a parasitic reflected light is detected underneath. Nine ns later, image E at

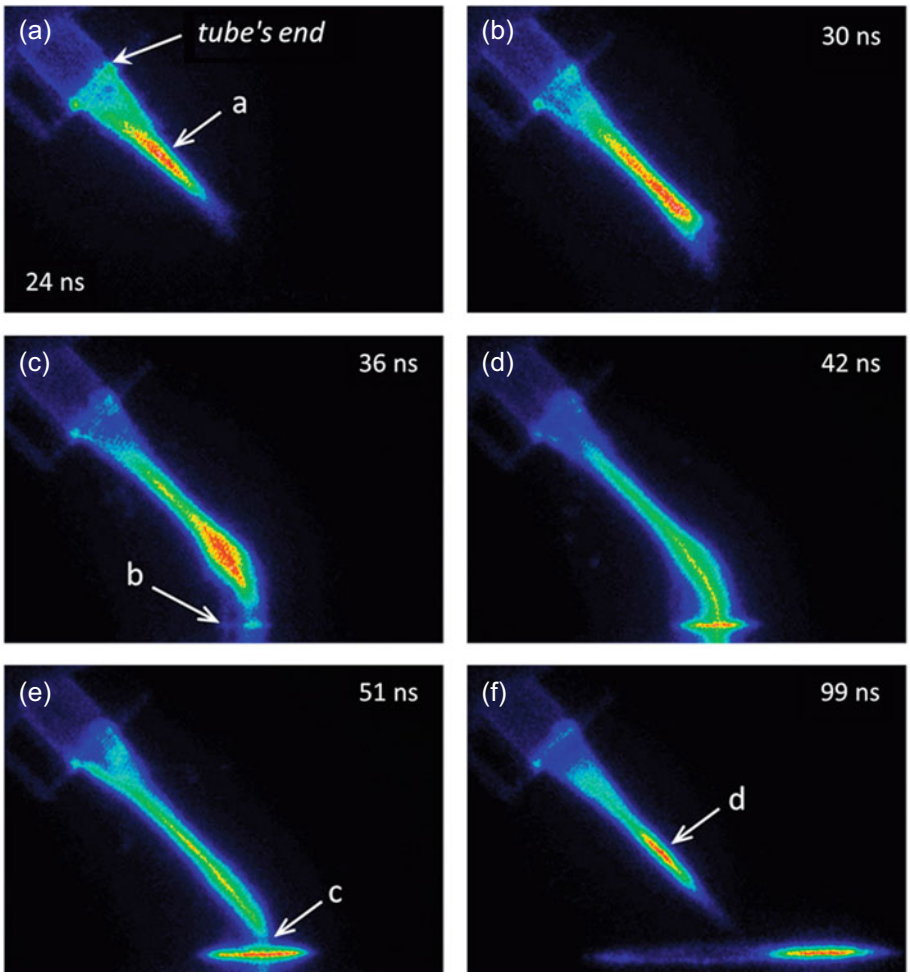


**Fig. 6.** Plasma emission captured by ICCD camera for an exposure time of 3 ns and accumulated over 100 successive HV pulses. Distance from nozzle to plate: 5.5 mm. Angle between the glass surface and the tube axis: 45°. Discharge parameters: 6 kV, 20 kHz. Argon flow rate: 0.25 L/min NTP. Time scale and arrows denoted "a", "b", "c", "d": please refer to text.

51 ns, the extension of the surface plasma in the observation plane symmetrically increases up to a width of about 3.2 mm; it is probable that part of the high intensity emission points on the surface correspond to the propagation of streamer heads in the direction of observation. At this time, a very low intensity area (indicated by arrow "c" in the image) can be seen between the plasma column created in the argon jet and the surface plasma. The last image, image F at 99 ns, shows that, 100 ns after the beginning of the streamers propagation outside the capillary tube, the plasma has spread downstream of the intersection point between the tube axis and the plate for about 3 mm (contrary to the plasma behaviour seen for an angle of 25° and the same gas flow rate – see top image in Fig. 5 – at this experimental conditions the distance traveled by the plasma surface is much shorter). On the surface, the width of the high intensity emission, 2.8 mm, is slightly lower than the width of the emission seen some 50 ns before on image E. It should be pointed out that a much less intense emission is now detected upstream, which could be due to streamers propagating from the plasma volume in the gas and impacting the surface, like those seen in Figure 5 (top image) for an angle of 25°, or could be due to some surface streamers coming from the intercept point mentioned above. The maximum intensity of the plasma emission in the gas phase, at a distance around 4.2 mm from the nozzle (indicated by arrow "d" in the image), is similar to the maximum intensity at the surface. However, a very low intensity (almost dark) area separates the intense plasma emission in the gas and the plasma on the surface. Images E and F highlight that a surface discharge, probably streamers, propagates while a plasma is maintained separately in the argon jet. We noticed also that this phenomenon becomes less important when the gas flow rate is increased up to 1 L/mn NTP, the dark area seen in images E and F disappearing almost completely for the highest flow rate studied.

## 5 Desorption of bibenzyl molecules

The results presented in the previous section are of great interest for the application of this argon plasma jet in the

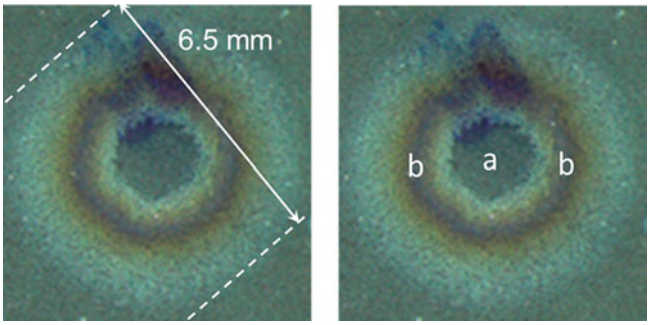


**Fig. 7.** The bibenzyl deposit on a glass surface after a perpendicular argon plasma jet treatment, showing the partial removal of the film. Discharge parameters: 6 kV, 20 kHz. Discharge running time: 10 s. Gas flow rate: 0.25 L/min NTP. Distance from nozzle to plate: 8 mm. Axis of the argon jet perpendicular to the plate. Areas denoted "a" and "b": see text.

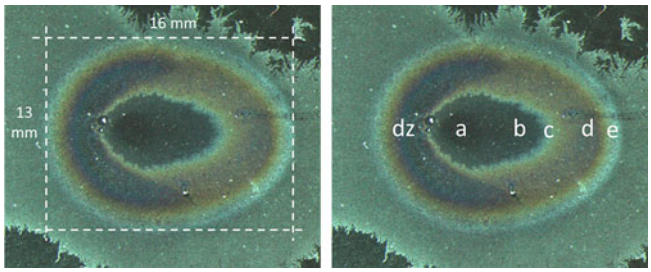


**Fig. 8.** The bibenzyl deposit on a glass surface after a non-perpendicular argon plasma jet treatment, showing the partial removal of the film. Discharge parameters: 6 kV, 20 kHz. Discharge running time: 60 s. Gas flow rate: 0.25 L/min NTP. Distance from nozzle to plate: 5 mm. Angle between the argon jet and the plate: 45°. Areas denoted "a", "b", "c", "d", "e", and "dz": see text.

desorption of heavy organic molecules. In order to study the viability of such application, the jet has been applied on glass plates in which bibenzyl molecules had been previously deposited. Firstly, it was verified that the argon jet alone (without the plasma) has no visible effect on the deposit. Besides, no organic molecule was detected by GC-MS in such experimental conditions.

### 5.1 Axis of propagation of the argon jet perpendicular to the glass plate

In Figure 7, it is shown a picture of the deposit after treatment by the plasma with the argon jet placed perpendicularly to the glass surface at a distance of 8 mm between the nozzle and the plate, and with a gas flow rate of 0.25 L/min NTP. Under these experimental conditions, the diffuse character of the plasma remained unchanged with respect to what is observed in Figure 4.

Although the discharge was turned on only for 10 s, a dark circle area (identified by "a" in Fig. 7) is observed in the center of the picture with a diameter of about 2 mm. This corresponds to a surface from where molecules were completely removed. The total visible area of the bibenzyl deposit affected by the plasma has a diameter of 6.5 mm, much greater than the argon jet section (maximum diameter of 1.7 mm). The surrounding area of the free from deposit central surface presents different optical aspects on concentric circles. The dark ring-shaped area ("b" in Fig. 7) could be related to the oxidation of molecules by reactive species (O, OH, $O_3$) produced by the plasma.

### 5.2 Axis of propagation of the argon jet non-perpendicular to the glass plate

Figure 8 shows the effect of the plasma on the deposit of bibenzyl while operating the plasma jet with an angle between the argon jet and the glass plate of 45°, and with all other experimental parameters being equal to those of Figure 6. The jet propagates from the left toward the right side of the image in Figure 8. The discharge running time was 60 ns in order to get an important visible effect on the layer of bibenzyl.

In this case, the impact of the plasma on the polluted plate presents an oval shape, which is the signature of the extension of the plasma on the surface. The dimensions of the area treated by the plasma are 16 mm for the largest axis and 13 mm for the smallest, again a much bigger surface than the intersection area between the argon jet and the glass plate.

The black circle in the center left of the picture in Figure 8 (identified by "a" in Fig. 8) shows a maximum removal of bibenzyl molecules. It is located at the intersection area between the argon jet axis and the glass surface. It is surrounded by a dark oval-shaped surface ("b" in Fig. 8) where few bibenzyl molecules are still present. Then, a bright narrow ellipse ("c" in Fig. 8) seems to be a bibenzyl area surrounded by a dark ellipsoidal ring-shaped area ("d" in Fig. 8), which is encircled by another bright ellipse ("e" in Fig. 8). The dark ellipsoidal crown corresponds probably to a zone where bibenzyl is oxidized on the surface. It is interesting to observe that in this dark area, a darker crescent-shaped zone ("dz" in Fig. 8) is located on the left side, resulting most likely from a greater oxidation activity or a higher surface density of oxidized molecules. Further studies are planned to correlate the treatment of the bibenzyl on the surface, using chromatographic measurements, and the spatio-temporal characteristics of the plasma determined by the ICCD camera.

More experiments were performed in order to get some insight on the effect of the gas flow rate and of the angle between the argon jet and the glass plate on the layer of bibenzyl. Results obtained with an angle fixed to 25° are given in Figure 9 for a gas flow rate of 0.5 L/min NTP and for a distance from the nozzle to the plate fixed to 5 mm. The area treated by the plasma has the same appearance as in Figure 8 for 45° and 0.25 L/min NTP, i.e., it shows concentric ellipsoidal crowns. However, the treated area is more elongated with a lower angle of impact and a higher gas flow rate. Measured dimensions are 15 mm for the largest axis, and 6.5 mm for the smallest. It should be noted that the latter value corresponds to the total

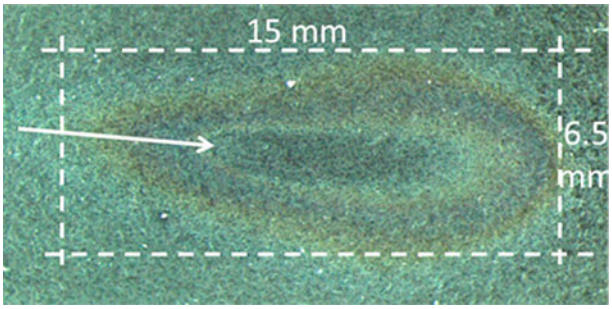


**Fig. 9.** The bibenzyl deposit on a glass surface after a non-perpendicular argon plasma jet treatment, showing the partial removal of the film. Discharge parameters: 6 kV, 20 kHz. Discharge running time: 30 s. Gas flow rate: 0.5 L/min NTP. Distance from nozzle to plate: 5 mm. Angle between the jet and the plate: 25°. The arrow shows the direction of propagation of the argon jet.

diameter of the surface treated by the plasma when the tube is placed perpendicularly to the plate (see Fig. 7). Greater surfaces are thus treated when the argon plasma jet is applied non-perpendicularly to the plate. In Figure 9, the dark area in the center of the image shows a maximum removal of bibenzyl molecules. The surrounding oxidation area is larger on the right side of the image, opposite to the arrival of the argon plasma jet (shown by the arrow).

Analyzes of gas samples taken above the plasma processing area show the presence of bibenzyl. Therefore, some of the initially deposited molecules are found intact in the gas phase. For a treatment time of 60 s, a jet angle of 45° at a distance of 5 mm from the plate, and for an argon flow rate of 0.25 L/min NTP (conditions for Fig. 8), an average mass of 20 ng of bibenzyl is detected. Further studies will be conducted to identify the by-products present in the oxidation areas.

## 6 Conclusion

The propagation of a DC-pulsed argon plasma jet through the surrounding ambient air, and its interaction with a glass plate placed on the argon jet trajectory, was studied by means of fast imaging. We observed that the dielectric surface plays an important role in the spatio-temporal characteristics of the plasma, allowing the creation of a diffuse plasma instead of a filamentary one for a short distance between the nozzle of the capillary tube and the glass plate, when the axis of the argon jet is placed perpendicular to the plate. Reducing the angle between the argon jet and the glass plate, and changing the gas flow rate allows to strongly modify the spatial extension of the plasma that develops on the surface. At low values of the gas flow there are many branchings in the gas phase connecting to the surface. Time-resolved measurements of the plasma emission showed that streamers begin to propagate following the tube axis, and thereafter deviate towards the surface from their main propagation path. Then, a surface discharge, probably streamers, propagates while a plasma is maintained separately in the argon jet.

This plasma jet shows interesting characteristics for desorption of low volatile organic molecules such as bibenzyl. A maximum removal of bibenzyl is located at the intersection area between the jet axis and the glass surface, surrounded by different zones where oxidation of the molecule is probably involved. It is interesting to note that the area treated by the plasma is much bigger than the intersection area between the argon jet and the glass plate, and that greater surfaces are treated when the argon plasma jet is applied non-perpendicularly to the plate. Finally, some of the initially deposited molecules are found intact in the gas phase.

Authors are grateful to V. Puech for valuable discussions. This work is supported by the French "Agence Nationale de la Recherche" under the PLASPAMS project (Grant No. ANR-2013-SECU-0002-03).

## References

1. X. Lu, M. Laroussi, V. Puech, Plasma Source. Sci. Technol. **21**, 034005 (2012)
2. S. Reuter, J. Santos Sousa, G. Stancu, J.-P. Hubertus van Helden, Plasma Source. Sci. Technol. **24**, 054001 (2015)
3. J. Winter, R. Brandenburg, K.-D. Weltmann, Plasma Source. Sci. Technol. **24**, 064001 (2015)
4. J.-S. Oh, O. Olabanji, C. Hale, R. Mariani, K. Kontis, J. Bradley, J. Phys. D: Appl. Phys. **44**, 155206 (2011)
5. M. Vandamme, E. Robert, S. Lerondel, V. Sarron, D. Ries, S. Dozias, J. Sobilo, D. Gosset, C. Kieda, B. Legrain, J.-M. Pouvesle, A. Le Pape, Int. J. Cancer **130**, 2185 (2012)
6. D. Graves, Phys. Plasmas **21**, 080901 (2014)
7. H. Paetzelt, G. Böhm, Th. Arnold, Plasma Source. Sci. Technol. **24**, 025002 (2015)
8. E. Szili, J.-S. Oh, S.-H. Hong, A. Hatta, R. Short, J. Phys. D: Appl. Phys. **48**, 202001 (2015)
9. B. Barwe, A. Stein, O. Cibulka, I. Pelant, J. Ghanbaja, T. Belmonte, J. Benedikt, Plasma Processes Polym. **12**, 132 (2015)
10. I. Onyshchenko, A. Nikiforov, N. De Geyter, R. Morent, Plasma Processes Polym. **12**, 466 (2015)
11. K. Duske, K. Wegner, M. Donnert, U. Kunert, A. Podbielski, B. Kreikemeyer, T. Gerling, K.-D. Weltmann, B. Nebe, R. Bader, Plasma Process. Polym. **12**, 1050 (2015)
12. G. Harris, A. Galhena, F. Fernandez, Anal. Chem. **83**, 4508 (2011)
13. J. Dalgleish, M. Wleklinski, J. Shelley, C. Mulligan, Z. Ouyang, R. Cooks, Rapid Commun. Mass Spectrom. **27**, 135 (2013)
14. S. Martínez-Jarquín, R. Winkler, Rapid Commun. Mass Spectrom. **27**, 629 (2013)
15. C. Douat, G. Bauville, M. Fleury, M. Laroussi, V. Puech, Plasma Source. Sci. Technol. **21**, 034010 (2012)
16. S. Huebner, J. Santos Sousa, V. Puech, G. Kroesen, N. Sadeghi, J. Phys. D **47**, 432001 (2014)
17. J. Garcia-Reyes, J. Harper, G. Salazar, N. Charipar, Z. Ouyang, R. Cooks, Anal. Chem. **83**, 1084 (2011)
18. Et. Es-Sebbar, G. Bauville, M. Fleury, O. Neveu, S. Pasquiers, V. Puech, J. Santos Sousa, in *Proceedings of the 8th International Workshop on Microplasmas, Newark, NJ, USA, 2015*