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PAPER

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Experimental and numerical investigation of the transient charging of a dielectric surface exposed to a plasma jet

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Abstract

This work investigates the dynamical charging of a surface under exposure of a non-equilibrium plasma jet at atmospheric pressure through a quantitative comparison between modeling and experiments. We show using mono-polar pulses with variable pulse duration and amplitude that the charging time (i.e. the time from impact of the ionization wave till the fall of the high voltage pulse) is a crucial element determining the plasma-surface interaction. This is done through direct measurements of the electric field induced inside the target using the optical diagnostic technique called Mueller polarimetry and comparison with the electric field calculated using a 2D fluid model of the plasma jet interaction with the target in the same conditions as in the experiments. When the charging time is kept relatively short (less than 100 ns), the surface spreading of the discharge and consequent surface charge deposition are limited. When it is relatively long (up to microseconds), the increased surface spreading and charge deposition significantly change the electric field to which the target is exposed during the charging time and when the applied voltage returns to zero.

Keywords: atmospheric pressure plasma jet, dielectric surface, plasma target interaction, electric field, ionization waves, Mueller polarimetry, surface charges

1. Introduction

Non-thermal atmospheric pressure plasma jets have been found to be a useful tool in a great variety of applications. This relates to the relatively simple and cheap design of the device which can be used to generate the repetitive ionization waves that allow for a chemical interaction with a target while the temperature of the heavy species remains low, thus preventing burning in addition to electrical damage.

A surface treatment by ionization waves is regarded to be beneficial due to a number of aspects that determine the plasma-surface interactions. There are radical species which are produced in the gas phase that get adsorbed on the surface, electric fields are induced in the substrate by (volume and surface) charged particles, there is a limited amount of heat delivered, and possible (UV) radiation by the discharge that could affect the interacting surface [1–3].

Applications for which non-thermal ionization waves have been used in the past to interact with substrates relate to surface functionalization of polymers, bio-medical treatment of living tissues, plasma assisted catalysis on powdered surfaces, and control of turbulent gas flows by inducing ionic

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winds by surface propagation of the discharge [4–6]. Atmospheric pressure plasma jets have been used for a great deal in bio-medical applications due to the precision where the interaction can be applied [7]. An array of jets can be used to treat simultaneously greater surfaces which is ideal for the treatment of plastics or fabrics.

Both the simplicity of the jet design and the numerous beneficial aspects of the plasma interaction have their downside as well. Ionization waves can be generated using various designs of the electrodes in addition to the gas flow rate (usually either helium or argon) and the voltage profile and amplitude which is applied. This causes a huge variety of plasma jets that are used throughout the world which makes it difficult to compare results that are obtained relating the surface treatment of similar substrates [8]. Secondly, the combined beneficial aspects of the surface interaction as mentioned above make it difficult to define a certain ‘*dosage*’ which is involved with the plasma-surface interaction [9].

In this work both problems will be addressed by investigating the charging of a surface by the impact of ionization waves, both experimentally and numerically. Positive mono-polar pulses are used with variable voltage amplitude and pulse duration to generate the discharges. It will be shown that the *charging time* of the surface, i.e. the time from impact till the fall of the mono-polar voltage pulse, allows for a greater control of the plasma-surface interaction.

In literature often a difference is emphasized between plasma jets operating at either radio-frequency or in the kHz-regime [10]. Within the kHz-regime however, the consequences of using either fast rising nanosecond pulses, microsecond mono-polar pulses, or continuous AC-sine waves is not often looked at.

The time-evolved periodical charging and discharging at the surface due to the impact of the ionization waves is examined experimentally by using electro-optic BSO targets. Their refractive index changes to the electric field induced by surface charges, according to the Pockels effect [11, 12]. This causes a birefringence that can be detected using the optical diagnostic technique called Mueller polarimetry [13].

The electric field that is measured and reported in this work and previous works characterizes the experienced electric field for a plasma jet interacting with high-dielectric constant materials at floating potential. When the dielectric constant is high, as will be shown in this work, the electric field inside the material is almost fully induced by surface charges, while volume charges play less a role. When the plasma jet interacts with materials of lower dielectric constant, as was shown in [14], both volume and surface charges are important for the electric field experienced by the target.

The numerical model describes the discharge dynamics in experimental conditions and is used to compare the spatio-temporal distributions of electric field with the experiments and provide further insight on those distributions. Jet-target interactions have been physically characterized in several works through either numerical simulations or experimental diagnostics. Recently, there is an effort to increase the understanding of discharge dynamics in jet-target interactions through comparisons between simulations and experiments.

Figure 1. The top-view of the experimental setup consisting of the Mueller polarimeter to examine the electro-optic BSO target under exposure of the non-thermal plasma jet either at the front side (A) or back side (B). The alignment of the plasma jet is along the *z*-axis with the BSO target in the *XY*-plane.

So far, these comparisons have evaluated tendencies and macroscopic parameters, such as discharge structure and velocity of discharge propagation [15–17]. In our last publication [14], a qualitative comparison between experiments and simulations has provided explanation for the difference between the rather high values of electric field measured in the jet plume and the rather low values measured inside a dielectric target. Furthermore, it has allowed to describe discharge dynamics and relate it with the electric field distributions. In this work, quantitative comparisons are performed on the temporally and spatially resolved electric field evolution, for various mono-polar applied voltage pulses in terms of amplitude and pulse duration.

The results section is discussed in three parts. The first shows the experimentally obtained electric field patterns together with the light emission during the plasma-target interaction for 6 kV pulses with variable pulse duration. This is compared to the numerical results in the second part by comparing the spatial and temporal profiles of the electric field and surface charge. The influence of the voltage amplitude on the charging of the surface and the electric field inside the target is shown in the last part.

2. Experimental setup

The Mueller polarimeter used throughout this work is shown in figure 1. The electro-optic BSO sample is examined at a 45° angle in transmission and it is placed in between the PSG (*polarizer state generator*) and PSA (*polarizer state analyzer*). The plasma jet impacts the target at normal incidence at a 10 mm distance. The plasma jet is in coaxial configuration with a stainless steel tube acting as powered electrode which is housed within a dielectric Pyrex tube (inner diameter 2.5 mm, outer 4 mm). A grounded ring is located 5 mm downstream from the end of the powered electrode and leaves

20 mm till the end of the capillary tube. Helium flows at 1 slm through the powered electrode into the open air environment. Various monopolar high voltage pulses (generator DEI PVX-4130) to generate the plasma are investigated with a constant rise and fall time of 50–60 ns and varying pulse duration from 230 ns to 960 ns and amplitude from 4 kV to 6 kV.

The PSG and PSA consist each of two ferro-electric liquid crystals with the retardance close to that of a quarter and half-wave plate, respectively. Externally, their orientation can be sequentially switched between two states which in total allows to capture 16 different intensity images at the iCCD camera. This forms a 4×4 intensity matrix which is the product of the three Mueller matrices of the PSG, the examined sample and the PSA, respectively. The Mueller matrices of the PSG and PSA are retrieved by performing the Eigenvalue Calibration Method [18, 19]. As a result the Mueller matrix of the examined sample can be recovered from the intensity matrix [20].

The Mueller matrix contains the optical properties of the sample relating to the diattenuation, depolarization and birefringence. The properties are further divided for the left/right circular polarization and linear polarization, both horizontal/vertical and along the two diagonals. This is customary to the Stokes description of polarized light. All these optical properties are entangled within the Mueller matrix and a decomposition is required to extract them. The logarithmic decomposition is best suited for our analysis since the sample is examined in transmission and the optical properties are accumulated homogeneously along the entire optical path through the sample [21, 22].

The two linear birefringences, in the horizontal/vertical optical system called $\Gamma_{0/90}$ and along the diagonal axes called $\Gamma_{45/135}$, that are obtained after applying the logarithmic decomposition are most important for this investigation since they are coupled to the externally induced electric field following the Pockels effect. Since the electro-optic sample is examined at an inclined angle of 45° , all the electric field components influence the birefringence [23]. This is different from the conventional case where the sample is examined at normal incidence and only the axial component (perpendicular to the surface) of the electric field induces a change of the refractive index. A second way to examine the axial field components is by using a different electro-optic crystal like Fe:LiNbO₃, as was shown in [23].

Two measurement configurations have to be used to separate the individual electric field components from each other, with the plasma jet impacting either from the front or the backside of the BSO crystal, respectively, as indicated in figure 1 by (A) and (B). The electric field components are retrieved using equation (1), with thickness $d = 0.5$ mm, wavelength $\lambda = 550$ nm, refractive index $n_o = 2.54$ and electro-optic constant $r_{41} = 4.8$ pm V⁻¹. Due to the refraction of the polarized light beam, the 45° inclination angle is changed to θ when the light propagates through the sample following Snell's law. This was not incorporated in [14, 23, 24] yet and affects the absolute values of the radial field components E_x and E_y . The axial field component E_z is unaffected by the examination angle of the crystal, since the

optical path through the material is $d^* = d / \cos(\theta)$

$$\begin{aligned} E_z &= \frac{1}{2} \frac{\Delta\Gamma_{A,0/90} - \Delta\Gamma_{B,0/90}}{2\pi d^* \lambda^{-1} n_o^3 r_{41} \cos(\theta)} \\ E_x &= \frac{1}{2} \frac{\Delta\Gamma_{A,0/90} + \Delta\Gamma_{B,0/90}}{2\pi d^* \lambda^{-1} n_o^3 r_{41} \sin(\theta)} \\ E_y &= \frac{1}{2} \frac{\Delta\Gamma_{A,45/135} + \Delta\Gamma_{B,45/135}}{2\pi d^* \lambda^{-1} n_o^3 r_{41} \sin(\theta) \cos(\theta)}. \end{aligned} \quad (1)$$

The electric field components can be detected time-resolved as was done in the previous works. This is possible with our Mueller polarimeter because the control of the liquid crystals and the acquisition of the iCCD camera is controlled simultaneously with respect to an external trigger event. This trigger relates to the generation of the high voltage pulse profile. As a result, all the acquired intensity images can be made on an identical time delay within the repetitive voltage profile and the charging of the material during the impact of the ionization waves can be followed in time. The polarimeter is fixed in location as well as the BSO target. To do the experiments, the jet is physically moved to the other side to do the second required measurement configuration. The distance towards the target, impact angle (90 deg) and initiation time of the plasma jet have been controlled carefully.

A more comprehensive discussion on the experimental setup is given in [24].

2. 1 Numerical setup

The model set-up is shown in figure 2. The geometry taken is the same as in the experiments. A dielectric quartz tube with relative permittivity of $\epsilon_r = 4$, length 3.3 cm (between $z = 1.0$ cm and $z = 4.3$ cm), internal radius $r_{in} = 1.25$ mm and outer radius $r_{out} = 2.0$ mm is used. Also, a dielectric BSO target with $\epsilon_r = 56$ is placed perpendicularly to the tube at 1 cm from the end of the tube. The target is defined as a cylinder of 2 cm radius and 0.5 mm thickness, set between $z = -0.5$ mm and $z = 0$. It is at a floating potential as in the experiments, and a grounded plane is set 10 cm behind it.

A powered ring electrode of inner radius 0.4 mm and outer radius 1.25 mm is set inside the tube between $z = 3.8$ cm and $z = 4.3$ cm and a grounded ring is wrapped around the glass tube between $z = 3.0$ cm and $z = 3.3$ cm, at a distance of 0.5 cm from the inner ring. Thus, unlike in our previous paper [14], in this work the electrode geometry and the target permittivity are the same in the model as in the experiments.

The computational domain is cylindrically symmetrical and the simulations have been carried out at atmospheric pressure and at $T = 300$ K. To simulate the discharge dynamics, we use as in [14] a 2D axisymmetric fluid model based on drift-diffusion-reaction equations for electrons, positive ions and negative ions and reaction equations for neutral species, coupled with Poisson's equation in cylindrical coordinates (z, r):

$$\frac{\partial n_i}{\partial t} + \vec{\nabla} \cdot \vec{j}_i = S_i \quad (2)$$

Figure 2. Side view schematics of the discharge set-up used in the simulations. The colour plot and the white curves show the O_2 spatial distribution in the He- O_2 mixture (percentage over a total of $2.45 \times 10^{19} \text{ cm}^{-3}$ gas density).

$$\vec{j}_i = (q_i/|e|)n_i\mu_i\vec{E} - D_i\vec{\nabla}n_i \quad (3)$$

$$\epsilon_0\nabla \cdot (\epsilon_r\nabla V) = -\rho - \sigma\delta_s \quad (4)$$

$$\vec{E} = -\vec{\nabla}V; \rho = \sum q_i n_i, \quad (5)$$

where the subscript i refers to each species and n_i , q_i , j_i , μ_i and D_i are the number density, the charge, the flux, the mobility and the diffusion coefficient of each species i , respectively. S_i is the total rate of production and destruction of species i by kinetic processes and by photoionization. V is the electric potential, $\vec{E}$ the electric field, e the electron charge, ϵ_0 the vacuum permittivity, ϵ_r the relative permittivity and δ_s the Kronecker delta (equal to 1 on the dielectric/gas interfaces). In this work, to simulate the discharge in the tube with an inner electrode and a grounded ring around the tube and the plasma jet interaction with a dielectric with a high permittivity ($\epsilon_r = 56$) we have improved the fluid model used in [14] and we have used the electron energy conservation equation and the local mean electron energy approximation [25]:

$$\frac{\partial}{\partial t}(n_e\epsilon_m) + \nabla \cdot \vec{j}_e = -|q_e|\vec{E} \cdot \vec{j}_e - \Theta_e \quad (6)$$

$$\vec{j}_e = -n_e\epsilon_m\mu_e\vec{E} - D_e\nabla(n_e\epsilon_m), \quad (7)$$

where $n_e = n_e\epsilon_m$ is the electron energy density, defined by the multiplication between the electron density and the electron mean energy, Θ_e represents the power lost by electrons in collisions and j_e is the flux of n_e by drift and diffusion.

It is important to point out that the numerical resolution of Poisson's equation (4) includes not only the geometrical Laplacian potential distribution, defined by boundary conditions, but also the net volume charge density ρ and the surface charge density σ contributions. At the surface of both the tube and the target, secondary emission of electrons by ion bombardment ($\gamma = 0.1$ for all ions) is taken into account. The surface charge density σ on the surface of the dielectrics is obtained by time-integrating charged particle fluxes through electric drift to the surface. These charges then remain immobile on the surface of the dielectrics. To model the experimental conditions, in which helium with impurities flows at 1 slm through the tube into air, the plasma model has been coupled with static flow calculations [14, 25]. As a first approximation, the model considers a flow of 1 slm of helium with 100 ppm of O_2 impurities flowing downstream into an O_2 environment. The spatial distribution of O_2 in the He- O_2 mixture obtained from the flow calculation is presented in figure 2.

In our previous works, the reaction scheme proposed in [26] has been used to describe the kinetics in the He- O_2 plasma. The scheme includes a total of 288 reactions with 5 positive ions, 5 negative species and 11 neutral species. In this work a reduced reaction scheme is used. The reduction consists on keeping only the 10 most important species in the μs timescales under study: e , O_2^- , He^+ , He_2^+ , O_2^+ , He , $He(2^3S, 2^1S)$, O , O_2 , $O_2(a_1\Delta_g)$. By excluding the reactions concerning the other 11 species, the scheme is reduced to 57 reactions. It has been verified that the discharge dynamics of propagation and thus the time of impact on the target and the radius of the discharge are not affected by the scheme reduction. Furthermore, the surface charge deposition on the dielectric target and the values of electric field inside the target, are affected in less than 10% by the scheme reduction. A study of the impact of the reduction is shown in another publication [27]. As a result of the chemistry reduction, the calculation time is reduced approximately by a factor 2.5.

The electron and electron energy transport parameters and the rate coefficients of electron-impact reactions are calculated with the electron Boltzmann equation solver BOLSIG+ [28], using the IST-Lisbon database of cross sections in LXCat [29, 30], and tabulated as functions of both the local gas mixture and the local mean electron energy ϵ_m . In each cell and at each timestep, each coefficient k is calculated for the local values of mixture and ϵ_m by linear interpolation between the upper and lower tabulated values and we obtain $k(\epsilon_m, \text{mix})$. For the photoionization model, we use the approach described in [31]. We assume that the VUV radiation emitted by excited helium species is absorbed by O_2 molecules of the admixture and ionizes them. The photoionization source term is proportional to the amount of O_2 admixture and we use $A_{ph} = \xi \times X_{O_2}$. In [25], we varied ξ in the range 10–1000 for different plasma jet/target

configurations. A small influence on the discharge structure and dynamics was observed. In this work, a value of 100 is used.

A finite volume approach and a cartesian mesh are used. The refinement taken requires a mesh of $n_z \times n_r = 4400 \times 370 = 1.628$ million points. The average computational time required for a simulation run in the μs timescale to obtain the results presented in this paper was of three days with 64 MPI processes on a multicore cluster ‘Hopper’ (32 nodes DELL C6200 bi-pro with two 8-core processors, 64 Go of memory and 2.6 GHz frequency per node). Further details on the numerical schemes and other characteristics of the simulations are given in [25].

2.2 Conditions for comparisons between experiments and simulations

As repetitive voltage pulses are used in the experiments ($f = 5$ kHz), memory effects are expected between the pulses. These can take the form of left-over charges in gas phase or on the dielectric surfaces or of other left-over species. The simulation results are dependent on the choice of initial conditions. For example, the velocity of discharge propagation towards the target and of spreading on the target surface has proven to be dependent on the choice of initial charge density on the target surface. Given the uncertainty on what the initial conditions in the simulations should be, we take into account a standard uniform initial preionization density $n_{\text{init}} = 10^9 \text{ cm}^{-3}$ of electrons and O_2^+ positive ions. However, no initial surface charges are considered on the dielectric surfaces. Moreover, an O_2 atmosphere is considered in the simulations, instead of an air atmosphere.

Due to the uncertainty in initial conditions and gas mixture, the velocity of discharge propagation and the time of discharge impact on the target in the simulations are different than those in the experiments. As in the experiments, the inner ring is powered by a positive mono-polar pulse that rises in 50 ns to a maximum V_p , and then remains constant. The pulse duration is variable and then the applied voltage decreases to zero in 50 ns. During the voltage rise and fall, a linear variation of the voltage is assumed. To compensate for the difference in time of impact, in the simulations the pulses are longer than in the experiments. The comparisons between experiments and simulations are performed for the same V_p and approximately the same charging time, rather than the same pulse duration. Short and long pulses are studied.

For short pulses, in the experiments the fall of the pulse is set to occur quickly after the time of impact of the discharge on the target. This cannot be done with the same accuracy as in the simulations since an exposure time of 25 ns is used and due to jitter of the ionization wave. In the simulations, the applied voltage starts decreasing at the time of impact.

For long pulses, the applied voltage potential remains high after the impact occurs for a certain time. In the experiments the applied voltage decreases to zero after 960 ns. In the simulations, the pulse duration is increased by approximately the difference in time of impact for each case. This is shown in table 1 which presents the times of impact

Table 1. Time of impact (ToI) of the discharge on the target in experiment and simulation for different V_p and pulse duration (PD). The pulse duration in simulations is extended to have comparable charging time as in the experiments for both relatively long and short interactions.

Experiment		PD	
V_p (kV)	ToI (ns)	long (ns)	short (ns)
6	166 ± 25	960	230
5	266 ± 25	960	330
4	441 ± 25	960	525
Simulation		PD	
V_p (kV)	ToI (ns)	long (ns)	short (ns)
6	320	1100	320
5	470	1200	470
4	750	1300	750

and the pulse durations (instant at which the applied voltage starts decreasing) in the experiments and in the simulations.

Afterwards the simulation results are shifted in time using table 1 to have the same time of impact as in the experiments. As a result, an excellent agreement between experiments and simulations is obtained on the physics and time dynamics of discharge-target interaction. No additional scaling or time shifting has been done for the experimental/numerical comparisons presented in this work.

3. Results and discussion

First, the plasma-surface interaction is examined for 6 kV mono-polar pulses with a relatively short pulse length. The pulse length is chosen for the *charging time* to be relatively short (i.e. the time from the impact at the surface till the fall of the high voltage pulse). The fall and rise time are around 50 ns and the frequency is 5 kHz. The fall of the pulse (pulse duration) starts at 230 ns. A minimum time duration of 150 ns is required, otherwise the ionization wave does not reach the surface and does not deposit charge meaning there would be no induced electric fields inside the material.

Figure 3 shows the time-resolved light emission during impact of the ionization waves and the resulting individual electric field components induced inside the targeted material due to deposited surface charges. The individual frames have been taken with an exposure time of 25 ns and for the light emission 500 frames have been accumulated. The delay mentioned in the figure is relative to the rise of the voltage pulse.

The plasma jet is located on the left-hand side and the ionization wave approaches the surface after 141 ns. A reflection of the light emission is visible as well since the light emission is captured at a 45° observation angle. The frames taken (25 ns exposure time) at a delay of 166 ns show that the

Figure 3. Light emission and the induced electric field components for short 6 kV pulses of 230 ns. The delay refers to the timing of the iCCD camera (25 ns exposure time) relative to the rise of the voltage pulse.

highest light emission is observed during the impact of the ionization wave with the surface and decreases rapidly afterwards.

The electric field patterns show a continuous charging of the material starting from the impact till the moment the high voltage pulse ends at 230 ns, hence defining the *charging time*. At a delay of 266 ns the electric field patterns have decreased already, most notably visible in the center of the axial E_z component. All the electric field components have decreased back to zero within 250 ns, indicating the time of removal/neutralization of the accumulated positive surface charges.

The maximum of the axial electric field lies in the center of the impact region with a value of 3.3 kV cm^{-1} . The radial E_x field component has its maximum of 3.6 kV cm^{-1} not in the center but 0.5 mm off-center, indicating the edge of the interaction area where charge deposition has occurred.

The dynamics of the charging and removal of charge at the targeted surface change when the pulse duration is increased from 230 till 960 ns. The light emission and electric field patterns obtained during the high voltage pulse (i.e. showing the charging) are shown in the top graph of figure 4, while the removal of deposited charges when the pulse falls back to zero is shown in the bottom figure. The observed electric field patterns/dynamics are representative for pulses longer than 500 ns. The impact occurs still at the same time as

with the short pulses but now since the high voltage pulse is maintained for longer times the surface interaction is altered.

Clear evidence is observed for the generation of stable surface streamers originating from the impact point moving radially outwards. A *star-shaped* pattern is seen which indicates that 8 streamers have occurred between 216 and 416 ns relative to the rise of the pulse at a fixed position relative to each other. If their position would not have been stable/repetitive, the high number of accumulations taken would have rendered the appearance of a ring structure instead. After 416 ns there is no more light emission observable from the surface streamers during the pulse and the electric field patterns also do not show a continuation of charge deposition.

The maximum value of the axial electric field component is 3.4 kV cm^{-1} . This matches the value observed for the relatively short pulse duration. However, the radial field components have increased in value, i.e. with maximum 5.3 kV cm^{-1} . This is a result of the increased total amount of charge that has been deposited since the surface area has increased. The fact that the axial field values are similar in amplitude suggests that the surface charge density is unaltered.

More experimentally obtained patterns of the electric field components have been obtained for variable pulse width/charging time at the surface. These have not been

Figure 4. Light emission and induced electric field for long 6 kV pulses of 960 ns. The delay indicates the timing relative to the rising of the mono-polar pulse. The top graph shows the plasma-surface interaction during the pulse and the bottom graph when the pulse ends.

added to this manuscript, since figures 3 and 4 are representative of the observed dynamics for short and long induced plasma-surface interactions, respectively. The results are available upon request to the authors.

As stated, the propagation of the surface streamers takes places within 200 ns after the impact occurs (and covers 3 mm radial distance) but the electric fields induced due to the deposited surface charges remain present over longer

Figure 5. Cross sections of the spatial distribution of the electric field magnitude E_t (on the left) and of the electron density n_e (on the right) from the simulations. Results for $V_P = 6$ kV and fall of pulse between 1100 ns and 1150 ns, at $t = 700$ ns.

timescales. It is only when the high voltage pulse is ended that the stability is broken, a second light emission event is observed and this is accompanied with the deposition of negative surface charges that induce electric fields of opposite polarity. The field changes first in the center of the impact region and then it spreads radially outwards but not as far as where the positive field is observed. The non-symmetrical patterns of charge deposition and removal could cause a (surface) memory effect that influences the sequential ionization wave impact. Unfortunately, the left-over surface charge density is too low to detect the electric field patterns on later time scales.

To examine the experimentally observed time-resolved charging of the surface in more detail, a quantitative comparison is made with the results from the numerical model. In the previous paper a qualitative comparison has been carried out. It has been shown that the discharge propagates with a high electric field at its head, around 20 kV cm^{-1} , associated to charge separation in volume in the discharge front. After the impact on the dielectric target surface, the discharge spreads radially and positive charge is deposited on the surface through electric drift of positive ions and secondary electron emission.

The same dynamics takes place in the simulations in the conditions of the current work. Figure 5 shows the spatial distribution of electric field magnitude E_t and of electron density n_e while the discharge is spreading on the target

surface, obtained through the 2D axisymmetric model. These results are taken at $t = 700$ ns for $V_P = 6$ kV and a long pulse which falls between 1100 and 1150 ns. The impact of the discharge on the target takes place 380 ns earlier, at $t = 320$ ns. It can be observed that the electric field is present mostly between the powered and grounded ring electrodes and on the edges of the plasma. In particular, a peak electric field is present on the front of the discharge, slightly above the target, allowing the discharge to continue propagating radially. We can also notice that a relatively high electron density remains in the plasma channel behind the propagation of the ionization wave.

As shown in [14], the electric field in the target is due to the charges deposited on the target surface and left-over volume charges. The surface charges are a result of the flux of charged species towards the target. When charges are present at the surface, a balance is created between the (positive) surface charges and the (positive) volume charges left in the plasma channel behind the head of the ionization wave. In [14], with a dielectric constant of 4, the experienced average electric field through the sample (located from $z = -0.05$ cm to $z = 0$ cm) had a maximum around 5 kV cm^{-1} .

In this work, we use the dielectric constant of BSO (i.e. 56) and consequently the experienced electric field inside the target is dominated by surface charges (over 90%) leaving the contribution from volume charges negligible. This is a direct consequence of Gauss's law at the boundary of the dielectric surface. As a result, an excellent quantitative agreement is obtained between the experimentally measured electric field experienced by the target and the numerically obtained electric field averaged through the target thickness. In addition, the numerical results are averaged over a 25 ns time interval making it comparable to the experimentally used exposure time.

This is visualized in figure 6 where the electric field inside the targeted material as shown in figure 3 is compared to the numerically obtained axial and radial components E_z and E_r , exclusively due to surface charges and averaged through the target thickness (0.5 mm) in similar conditions. A horizontal cross-section starting from the center of the impact point towards the right is taken for the axial E_z component and the radial E_x component (numerically this is E_r) and shown for different time delays relative to the rise of the mono-polar pulse. Additionally to the electric field profile is also shown the (numerical) surface charge density profile.

Before the impact of the ionization wave, shown by subfigure (A), the electric fields are zero since no detectable surface charges are present. As a result, the electric field should be zero and the experimentally shown profiles are representative of the noise levels/uncertainty. This detected level of noise before impact is used as uncertainty in the following graphs. The experimental uncertainty/noise for the radial field component is slightly larger than for the axial component. This is because the detected birefringent signal Γ depends more strongly on E_z than on E_r due to the $\sin(\theta)$ and $\cos(\theta)$ terms in equation (1).

Figure 6. Comparison between the experimentally and numerically obtained radial profile of electric field (kV cm^{-1}) induced inside the target at different time delays relative to the rise of the mono-polar pulse: (A) time delay = 141 ns and for (B)–(H), respectively, (166, 191, 216, 241, 266, 291, 316) ns. The numerical time delay is shifted by 160 ns. The 6 kV high voltage pulse remains high till 230 ns. Also the (numerical) surface charge density is shown in red (nC cm^{-2}). The distance indicates the horizontal cross-section starting in the impact point.

After the impact, while the high voltage potential remains at 6 kV, the total charging of the target continues as shown by subfigures (B)–(D). Both experimentally and numerically it is observed that there is a radial front that propagates approximately 0.5 mm away from the center. The radial field patterns are highest around this same point. When the high voltage potential starts decreasing at 230 ns, negative charge deposition takes place and the electric field profiles decrease since the surface charge density returns to zero. This is because there is a redistribution of the potential lines which causes the electric field in the gas phase to be reversed. This occurs faster in the center of the impact point than at a 0.5 mm radial distance.

When a long pulse duration is used, as shown experimentally in figure 4, the surface dynamics are clearly different and the appearance of surface streamers is evident. They originate not immediately but after the initial impact where a more homogeneous axial electric field pattern is observed. The radial surface streamers cannot be described by the 2D-axisymmetric model used throughout this work, making it pointless to compare spatial profiles for longer pulses as done in figure 6. However, the electric field induced in the center of the impact point can still be compared especially during the earlier stages of the plasma-surface interaction.

Figure 7 shows the amplitude of the induced electric field in the center of the impact region as a function of time for $V_p = 6$ kV and three different pulse durations. Additionally to the 230 ns and 960 ns pulse, also the results of a 470 ns pulse are shown and compared between experiments and simulations. The moment of impact is always at 166 ns in the experiments (and 320 ns in the simulations) while the maximum (axial) electric field of 3.3 kV cm^{-1} is reached at 191 ns relative to the rise of the voltage pulse. The initial charging

Figure 7. Temporal evolutions of $E_{zav\sigma}$ at $r = 0$ from the experiments and from the simulations for different pulse lengths with a fixed amplitude of 6 kV. The numerical results are shifted in time to show the equal charging time between experiments and simulation.

during impact of the material is the same for all voltage pulses, both experimentally and numerically when the proper time shift is taken, as explained in section 2.2. After this first stage, the surface interaction starts to be dependent on the pulse duration and thus the charging time of the sample.

For the relatively short pulse of 230 ns the electric field returns to zero since negative charges are deposited because of the reversal of electric field in the gas phase when the potential redistributes at the end of the pulse. For longer pulses this does not happen right after the impact since the applied voltage first remains high and a second stage of the plasma-surface is initiated. Experimentally, the surface

streamers start to be observed moving radially outwards and creating the stable star-shaped pattern. Numerically, the ionization front also moves radially outwards but since it cannot represent the asymmetrical streamers we focus on what happens on the axis in the center of the impact point.

Figure 7 shows that for the 470 and 960 ns pulses the electric field decreases much slower than for the 230 ns pulses and it stagnates at an electric field of approximately 1 kV cm^{-1} . This is caused by a stabilization between the deposited surface charges and the volume charges left behind the head of the ionization wave in the plasma channel (while the high voltage remains at 6 kV). Thus, the longer the pulse is and the radial spreading of the discharge continues, the longer is the decrease of the average electric field by surface charges at the center before the end of the pulse.

The third and final stage of the plasma-surface interaction is initiated when the mono-polar pulse comes to an end at, respectively, 470 or 960 ns. It was shown in figure 4 that a second light emission event is observed experimentally (which is not observable with the smaller 230 ns pulses) at the (negative) fall of the high voltage potential. It is caused by the electrical redistribution that propagates rapidly from the powered electrode towards the target. This propagation, as evidenced by the light emission, is much faster than the propagation of the ionization wave during the pulse because of the high level of charge density in the still conductive channel. The electric field is reversed in the gas phase which means that negative charges are deposited. This lowers the experienced electric field in the sample.

The amount of positive charges in the center of the impact point had already decreased during stage two. This allows for a local surplus of negative charges to be observed, causing an overall negative electric field in the center of the impact region. This is most profoundly seen for the 960 ns mono-polar pulse. The recombination of the negative charges that remain takes the longest of all the observed dynamics. The amplitude of the negatively observed electric field increases with the pulse duration. This is caused by the increase of total amount of positive charges that is deposited during the pulse. With long pulse duration the surface propagation of the ionization wave is increased which enhances the total interaction area and thus the total (positive) charge deposited. This means more negative charges are attracted when the field reversal occurs at the end of the pulse.

Another parameter of control of the pulsed jet-target interaction is the magnitude of the applied voltage. Previous studies of positive streamer-like discharges without target [32–34] have shown that the increase of the applied voltage leads to the increase of discharge radius and propagation velocity. However, the values of electric field in the ionization front and electron density in the plasma are reported to be independent of the applied voltage in [32] and to decrease with the applied voltage in [33]. Here we evaluate the influence of the applied voltage on the jet-target interaction and the electric field inside the target.

Figure 8 presents the temporal evolutions of $E_{zav\sigma}$ averaged inside the target at $r = 0$ from both the experiments and the simulations for different magnitudes of applied voltage

Figure 8. Temporal evolutions of $E_{zav\sigma}$ at $r = 0$ from the experiments and from the simulations for short pulses with a voltage amplitude of 4, 5 and 6 kV. The applied voltage starts to decrease (with a 50 ns fall time) at, respectively, 525, 330 and 230 ns for experiments and 750, 470 and 320 ns in the simulations. The numerical results are shifted in time as explained in section 2.2 to show the equal charging time.

V_p : 4, 5 and 6 kV. In both cases, only short pulses are considered. In the experiments, the fall of the pulse is set closely after the time of impact of the discharge on the target. In the simulations, the applied voltage starts decreasing at the time of impact and takes 50 ns to reach zero. The times of fall of the applied voltage are noted in the figure caption. The numerical results are shifted in time as described in section 2.2.

The three cases represented in figure 8 show an excellent agreement between experiments and simulations. Even though the discharge radius and velocity are higher with higher V_p , the fall of the applied voltage at the time of impact limits the radial expansion of the discharge on the target for every case. Therefore, the jet-target interaction is very similar in all cases, as is the increase of $E_{zav\sigma}$ at $r = 0$ up to approximately 3.5 kV cm^{-1} and its decrease to zero in approximately 300 ns. We should notice that there is no negative charge deposition after the decrease of $E_{zav\sigma}$ to zero in any case.

Then, in figure 9 we assess the temporal evolutions of $E_{zav\sigma}$ at $r = 0$ for three cases of applied voltage with longer pulses. In the experiments, the pulses start decreasing at 960 ns. For the 4 and 5 kV pulses an exposure time of 100 ns is used instead of 25 ns, which did not effect the results. In the simulations, the pulses are longer but shifted in time, in order to have approximately the same charging time as in the experiments, which is verified in the figure. The times of fall of the applied voltage are noted in the figure caption.

It can be observed in figure 9 a good agreement between numerical and experimental results. In the simulation results, the maximum of $E_{zav\sigma}$ at $r = 0$ is around 3.5 kV cm^{-1} , the same as with the short pulses in figure 8. In the experiments, that is also the case for $V_p = 5$ and 6 kV. However, with $V_p = 4 \text{ kV}$, $E_{zav\sigma}$ at $r = 0$ in the experiments rises higher, up

Figure 9. Temporal evolutions of $E_{zav\sigma}$ at $r = 0$ from the experiments and from the simulations for long pulses with a voltage amplitude of 4, 5 and 6 kV. The applied voltage starts to decrease (with a 50 ns fall time) at, respectively, 1300, 1200 and 1100 in the simulation, while in the experiments (for all voltages) the pulse falls at 960 ns. The numerical results are shifted in time as indicated to show the equal charging time.

to 5 kV cm^{-1} . Moreover, the maximum is attained only around 300 ns after the time of impact of the discharge on the target.

This result suggests that in that particular case the positive charge deposition continues for 300 ns close to the point of impact, before the discharge spreads radially and the positive charge at the center starts decreasing. That is not the case in the simulation results and in the other two experimental results, in which cases the radial spreading starts shortly after the impact, and thus $E_{zav\sigma}$ at $r = 0$ starts decreasing faster. Yet, in both experimental and numerical results it is noticeable that $E_{zav\sigma}$ at $r = 0$ between its maximum and the end of the pulse decreases faster for higher applied voltage. As a result, $E_{zav\sigma}$ at $r = 0$ remains higher for lower V_p , which is counterintuitive. Furthermore, after the fall of the voltage pulse, higher V_p leads to more negative $E_{zav\sigma}$ at $r = 0$, in both simulations and experiments.

The evolution of electric field and charge deposition with long pulses and different voltage amplitude is explained by the same mechanisms that explain the results in figure 7. The radial spreading is faster and further with higher voltage amplitude. This means that there is more positive charge deposition at greater radial positions. As a result, more negative charge deposition takes place at the center of the impact point until reaching an electrical quasi-equilibrium between the plasma and the target both during phase two and after the fall of the pulse.

4. Conclusions

The comparisons between experiments and simulation show that the charging time of the target is a crucial parameter in describing the dynamics that determine the electric field induced by the plasma inside the (high permittivity) material. The charging time is determined from the moment of impact till the high voltage pulse ends.

The voltage amplitude is an important parameter as well. Firstly, it changes the velocity of the ionization waves. This

changes the charging time at the surface if a constant pulse duration is used. When the pulse duration is decreased to keep constant charging time with higher voltages, the experienced electric field during the plasma-surface interaction is less influenced by the voltage amplitude. Only a small decrease in electric field amplitude is observed. Secondly, when the pulse duration is longer, the surface propagation is dependent on the applied voltage and thus it influences the amount and area of charge deposition and recombination.

A very good agreement between simulations and experiments is obtained when the same geometry (coaxial jet configuration with an inner powered electrode and outer grounded ring) together with the high dielectric permittivity target are considered in the model and in the experiments. With a permittivity of the target equal to 56%, 90% of the electric field in the target is induced by the temporarily deposited surface charges.

Naturally, still differences are present between the model and the experiment. In the experiments when a long pulse duration is applied, stable surface streamers are observed that induce a star-shaped pattern. This pattern is observed even though a high number of acquisition events is averaged to obtain the result. The occurrence of the constant star-shaped pattern gives an indication that self organization occurs when the stable non-branching ionization wave impacts the surface. Left-over surface charges from the previous voltage cycle could play a role in this pattern formation. The numerical model has a two dimensional axis-symmetrical configuration, meaning that the surface branching/pattern formation cannot be described. That is why we focused in this paper on the electric field values induced in the center of the impact point. The stable surface streamers are initiated not from the center but from a ring located at a 0.5 mm radial distance.

The second main difference between the model and the experiments is the off-set of the propagation velocity of the ionization waves. In the model the velocity is slightly lower most likely because of memory effects, such as residual charge deposition at the dielectric surfaces. This causes the impact moment at the target to be delayed. By imposing an extension of the mono-polar pulse duration equal to this delay, a comparable charging time is studied in the model for the plasma-surface interaction compared to the experiments.

These observations and conclusions related to the charging time controlled by the pulse duration and the voltage amplitude are a first step to allow comparison between different jet designs, operating parameters and treatments of targets and go closer to a future definition of what a possible defined ‘dosage’ delivered by the plasma jet must entail.

Mono-polar pulses are an ideal tool to control and study the plasma-surface interaction. For pulses with a relatively short duration, the temporarily deposited surface charges induce electric field but since the pulse ends quickly the ionization wave cannot spread at the surface and the induced electric field decreases within 300 ns. This is caused by the reversal of the electric field in the gas phase when the potential drops and as a result negative charges are deposited at the surface right after, which limits the total interaction time. This is related to ns-pulsed systems since then also the charging time of the surface is limited.

When the pulse duration is longer, the ionization waves are able to spread along the surface (which depends on the voltage amplitude) after the impact since the potential is maintained. As a result, the surface charges decrease much slower during the pulse and stay present until the high voltage pulses come to an end and the reversal of the electric field causes negative charge deposition. Then, since the ionization wave has spread radially further and thus charged a larger area of the material, an overshoot of negative charges can be deposited in the center of the impact region at the fall of the pulse, especially when higher voltage amplitudes are used.

The surface dynamics observed for longer pulses become more comparable to AC driven plasma jet systems. Of course the consequences of a time variable potential of the plasma channel compared to a constant potential cannot be overlooked, but it is notable that for both systems charges are deposited during the impact of the ionization wave and remain there for longer times. For AC systems the removal is initiated when the polarity of the potential changes and the negative half period begins [11], while for the mono-polar pulses positive charges remain present until the potential falls back to zero.

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References

- [1] Laroussi M and Leipold F 2004 *Int. J. Mass Spectrom.* **233** 81–6
- [2] Weltmann K D, Kindel E, Brandenburg R, Meyer C, Bussiahn R, Wilke C and von Woedtke T 2009 *Contrib. Plasma Phys.* **49** 631–40
- [3] Graves D B 2012 *J. Phys. D: Appl. Phys.* **45** 263001
- [4] Noeske M, Degenhardt J, Strudthoff S and Lommatzsch U 2004 *Int. J. Adhes. Adhes.* **24** 171–7
- [5] Kong M G, Kroesen G, Morfill G, Nosenko T, Shimizu T, van Dijk J and Zimmermann J L 2009 *New J. Phys.* **11** 115012
- [6] Moreau E 2007 *J. Phys. D: Appl. Phys.* **40** 605–36
- [7] Stoffels E, Flikweert A J, Stoffels W W and Kroesen G M W 2002 *Plasma Sources Sci. Technol.* **11** 383–8
- [8] Lu X, Laroussi M and Puech V 2012 *Plasma Sources Sci. Technol.* **21** 034005
- [9] Gidon D, Graves D B and Mesbah A 2017 *Plasma Sources Sci. Technol.* **26** 085005
- [10] Kim D B, Rhee J K, Gweon B, Moon S Y and Choe W 2007 *Appl. Phys. Lett.* **91** 151502
- [11] Slikboer E, Guaitella O and Sobota A 2016 *Plasma Sources Sci. Technol.* **25** 03LT04
- [12] Slikboer E, Garcia-Caurel E, Guaitella O and Sobota A 2017 *Plasma Sources Sci. Technol.* **26** 035002
- [13] Slikboer E, Sobota A, Guaitella O and Garcia-Caurel E 2018 *J. Phys. D: Appl. Phys.* **51** 025204
- [14] Viegas P, Slikboer E, Obrušnik A, Bonaventura Z, Sobota A, Garcia-Caurel E, Guaitella O and Bourdon A 2018 *Plasma Sources Sci. Technol.* **27** 094002
- [15] Ning W, Dai D, Zhang Y, Han Y and Li L 2018 *J. Phys. D: Appl. Phys.* **51** 125204
- [16] Lazarou C, Anastassiou C, Topala I, Chipera A S, Mihaila I, Pohoata V and Georgiou G E 2018 *Plasma Sources Sci. Technol.* **27** 105007
- [17] Ji L, Yan W, Xia Y and Liu D 2018 *J. Appl. Phys.* **123** 183302
- [18] Compain E, Poirier S and Drevillon B 1999 *Appl. Opt.* **38** 3490–502
- [19] Macías-Romero C and Török P 2012 *J. Eur. Opt. Soc. Rapid Publ.* **7** 12004
- [20] Garcia-Caurel E, Ossikovski R, Foldyna M, Drévillon B and Martino A D 2013 *Ellipsometry at the Nanoscale* (Berlin: Springer) (https://doi.org/10.1007/978-3-642-33956-1_2)
- [21] Azzam R M A 1978 *J. Opt. Soc. Am.* **68** 1756–67
- [22] Ossikovski R 2011 *Opt. Lett.* **36** 2330–2
- [23] Slikboer E, Sobota A, Guaitella O and Garcia-Caurel E 2018 *J. Phys. D: Appl. Phys.* **51** 115203
- [24] Slikboer E T 2018 Investigation of plasma surface interactions using mueller polarimetry *PhD Thesis* Technische Universiteit Eindhoven École Polytechnique, Université Paris-Saclay
- [25] Viegas P 2018 Electric field characterization of atmospheric pressure Helium plasma jets through numerical simulations and comparisons with experiments *PhD Thesis* École Polytechnique University Paris-Saclay, France
- [26] Liu D X, Rong M Z, Wang X H, Iza F, Kong M G and Bruggeman P 2010 *Plasma Process. Polym.* **7** 846–65
- [27] Viegas P and Bourdon A 2019 *Plasma Chem. Plasma P.* submitted
- [28] Hagelaar G J M and Pitchford L C 2005 *Plasma Sources Sci. Technol.* **14** 722–33
- [29] Pancheshnyi S, Biagi S, Bordage M, Hagelaar G, Morgan W, Phelps A and Pitchford L 2012 *Chem. Phys.* **398** 148–53
- [30] IST 2018 IST-Lisbon database, www.lxcat.net, retrieved on 1 June 2018
- [31] Bourdon A, Darny T, Pechereau F, Pouvesle J M, Viegas P, Iséni S and Robert E 2016 *Plasma Sources Sci. Technol.* **25** 035002
- [32] Babaeva N Y and Naidis G V 1996 *J. Phys. D: Appl. Phys.* **29** 2423–31
- [33] Kulikovsky A A 1998 *Phys. Rev. E* **57** 7066–74
- [34] Naidis G V 2012 *J. Appl. Phys.* **112** 103304