Increase of electric field on streamer/surfaces contact area with using external electrode

I V Schweigert$^{1,2}$, S Vagapov$^1$, L Lin$^2$, M Keidar$^2$

$^1$Khristianovich Institute of Theoretical and Applied Mechanics, 630090, Institutskaya str 4, Bld. 1, Novosibirsk, Russia
$^2$George Washington University, D.C. 20052, 2121 I St NW, Washington, USA

ischweig@yahoo.com

Abstract. Nowadays, a low-temperature atmospheric plasma jets are widely used in the anticancer therapy. Enhancement of electric field near the surface is one of possible ways to increase an efficiency of plasma jet treatment. Another mechanism of plasma jet’s influence on living cells is generating reactive oxygen nitrogen species (RONS). In this work effect of presence of external biased electrode was studied. Electric fields near the dielectric surface and zone of interaction were much higher than that near the conductive one due to accumulation of charge on the surface. The evolution of ionization rate near the surface was studied. It was observed experimentally and in the simulation that there are two main stages of evolution – the stage of fast ionization which is few hundreds of ns long and the stage of slow ionization with characteristic time of $\sim 10$ $\mu$s. A 0D modelling was applied in the boundary layer near the surface in order to obtain mixture composition of RONS after several pulses.

1. Introduction
Cold atmospheric-pressure plasma jets attract attention of researchers from various areas of science for about few decades. The reason of such interest is a wide spectrum of applications of plasma jets, especially in medicine [1] as a promising tool for cancer treatment. Due to presence of strong electric field, high energy electrons, ions and metastable atoms and molecules in the head of the streamer a lot of various reactive oxygen nitrogen radicals (RONS) are being generated in the vicinity of the surface. Also it is believed that plasma jets can influence on cells due to delivery of electric field to the biological tissue and this, along with RONS, may have a synergetic effect.

In this work we provided increasing of electric field with ring external electrode installed between tube’s nozzle and the surface. The main streamer’s characteristics such as rate of ionization and electric field were investigated with and without external electrode. Streamer’s interaction with conductive and dielectric surfaces was also studied. Using obtained characteristics of the streamer above the surface we implemented 0D modelling to estimate amounts of RONS generated during plasma jet treatment.

2. Experimental setup and model description
Plasma jet and computation domain are illustrated in figure 1. Plasma is generated between two electrodes powered with sinusoidal voltage at 12.5 kHz by feeding 13.5 sl/min helium through...
dielectric tube. The tube has nozzle diameter of 0.46 cm. The copper or dielectric targets are placed at 3 cm from tube’s nozzle. External electrode is placed at 1 cm from tube’s nozzle. Its external and internal radii are 0.65 cm and 0.185 cm, respectively. The propagation of plasma bullet is recorded by an Andor iStar ICCD camera with exposure time – 10 ns. The experimental setup is described in details in [2].

2D modelling includes Poisson’s equation, continuity equations for electrons, ions and mean electron energy. Molar fractions of He, O₂, N₂, H₂O during the mixing of helium flow with ambient air were pre-calculated using ANSYS Fluent. Transport coefficients in the continuity equations for electrons and their energy were calculated from the solution of Boltzmann’s equation. For ions transport coefficients were given by the next analytical expressions:

\[
\mu_i = \frac{3e}{16n_e \sigma_i} \left( \frac{\pi}{m_i kT} \right)^{1/2} \left( 1 + \frac{9\pi^2 eE}{512 n_g kT \sigma_i} \right)^{-1/2}
\]

\[
D_i = \mu_i kT
\]

where \( \mu_i \) is the mobility of ions, \( D_i \) is the coefficient of diffusion of ions, \( e \) is an elementary charge, \( n_e \) is the concentration of electrons, \( k \) is the Boltzmann constant, \( T \) is the gas temperature, \( E \) is the electric field, \( m_i \) is the ion’s mass and \( \sigma_i \) is its transport cross-section.

The coefficient of recombination rate \( \beta \) for helium at atmospheric pressure and 300K is \( \beta = 2 \times 10^{-7} \) cm³/s [3]. The secondary emission coefficient \( \gamma \) from copper target is 0.02 [4]. For the dielectric surface \( \gamma \) was taken 0 and 0.1.

The calculation domain has a cylindrical symmetry with \( R_{\text{max}} = 5 \) cm and \( H_{\text{max}} = 5.5 \) cm. Three values of voltage amplitude \( U_0 \) were investigated – 3.9 kV, 5 kV and 7 kV.

Figure 1. Computational domain and plasma jet.

Figure 2. Image of the streamer (a), calculated ionization rate in cm⁻³/s for \( U_0 = 5 \) kV (b), spatial distributions of electron energy in eV (c) and electric field in kV/cm (d) in linear scale.
3. Results and discussion

3.1. Main streamer characteristics
When voltage on the powered electrode reaches threshold value wave of ionization starts to propagate from the anode over the dielectric tube. The optical emission (see figure 2 (a)) indicates an approximate radius of the streamer channel, which is essentially smaller than the radius of a wider part of the tube. It is seen from figure 2 (b) that streamer has toroidal shape with external and internal radii of 0.185 cm and 0.14 cm, respectively. Figures 2 (c) and (d) illustrate spatial distributions of electron energy and electric field. The maximum values are 11 eV and 17 kV/cm, respectively. The electron density in the streamer head is $2 \times 10^{12}$ cm$^{-3}$.

3.2. Effect of external electrode
As was mentioned above the external ring electrode was maintained at 1 cm from tube’s nozzle. We applied positive and negative (+1.5 kV and -1.5 kV) voltages $U_r$ to the ring and studied a variation of streamer characteristics.

Figure 3 illustrates spatial potential distributions for cases with +1.5 and -1.5 kV at the moment when the streamer arrives to the surface. In the case of negatively biased ring there is larger electric field near the surface. For both cases a spot of ionization has the shape of the ring of 0.7 mm wide with external radius of 0.19 cm. The rate of ionization exponentially decays to the center of the ring as it can be seen in figure 3 (c).

Evolution of the ionization rate near the surface for various cases is illustrated in figure 4. It can be seen that for $U_0 = 5$ kV and with the external electrode ionization rate was larger by factor of two. Electric field in this case was also larger by 30%.

For all cases the ionization rate has the same character of evolution and two stages can be distinguished. In the first one which is 100-150 ns long ionization rate reaches its maximum when the streamer approaches the surface. The second one lasts approximately 10 $\mu$s and it is characterized by much less intensive ionization.

This evolution of ionization agrees with experimental observations. In figure 5 experimental images of streamer over the Cu-plate at various moments of time are presented. The first image shows much more intensive optical emission what corresponds to the first stage. The next images show lower optical emission which remains constant for the next ~10 $\mu$s which can indicate the occurrence of the second part.

![Figure 3](image_url)  
**Figure 3.** Spatial potential distribution in kV for $U_r = -1.5$ kV (a) and 1.5 kV (b) for $U_0 = 5$ kV. Ionization rate (c) with colour palette $(0.01-2) \times 10^{22}$ cm$^{-3}$ s$^{-1}$ for $U_r = -1.5$ kV.
Figure 4. Evolution of ionization rate near the surface for $U_0 = 5$ kV, $U_r = -1.5$ kV (a), for $U_0 = 5$ kV, no ring (b) and for $U_0 = 3.9$ kV, no ring (c).

Figure 5. Images of the streamer near the metal surface

3.3. Streamer interaction with dielectric surface
Streamer interaction with metal and dielectric surfaces with various value of $\varepsilon/d$ (d is dielectric’s thickness) and various configurations already was studied in [5, 6].

In this work we studied streamer interaction with dielectric with $\varepsilon = 10$ in the presence of negatively biased ring electrode. A dielectric which is 0.3 cm thick was placed on the grounded metal plate. Spatially non-uniform surface charge accumulates when streamer approaches to the surface. In the case with $\gamma = 0.1$ ionization wave starts to propagate along the surface. In the case with non-emissive surface there are not enough electrons crossing the leading front of the ionization wave to maintain its propagation.

Axial and radial electric field components $E_z$ and $E_r$ over the emissive surface are illustrated in figure 6. $E_z$ can achieve 80 kV/cm near the surface. The radial component $E_r$ appears due to non-uniform surface charge distribution. Its maximum is 22 kV/cm. Due to presence of $E_r$ the value of electric field is much higher than that over the conductive surface. Inside of dielectric, the $E_z$ ranges...
Figure 6. Spatial distribution of electric field (a) and (b) in V/cm over an emissive dielectric surface for $U_0 = 5$ kV and $U_r = -1.5$ kV.

from 10 kV/cm to 6 kV/cm and the $E_r$ varies from 12 kV/cm to 4 kV/cm. It is seen that close to the center the $E_r$ changes its sign.

4. 0D model of plasma chemistry

We use 0D model for calculation of plasma enhanced chemical reactions in the thin boundary layer near the surface where speed of the gas flow is almost zero. So there is no need to consider the convection of species.

Kinetic scheme consists of 872 chemical reactions and 98 corresponding components. The scheme includes reactions of ionization (including Penning ionization), dissociation, recombination, charge exchange, excitation and quenching of electronic, vibrational and rotational levels. 190 of them are electron-impact processes. Rate coefficients were taken from literature for ion-ion, neutral-ion and neutral-neutral reactions and were calculated from the solution of Boltzmann equation using cross-sections for electron-impact processes and a mixture composition. Boltzmann equation has been solved periodically with BOLSIG+ solver in order to obtain tables of rate coefficients which were interpolated for current value of the electron temperature.

2D simulation and experiment revealed that after streamer reached the surface during $t_s = 1-10$ µs (corresponding to the positive cycle of voltage) rates of ionization and excitation are almost constant. As seen from figure 5, the light emission spot has the same intensity for several µs after the streamer approaching the surface. In our simulation of plasma chemistry, we chose $t_s = 5$ µs and assume that the electron concentration and temperature are constant. For $t_s > 5$ µs the ionization and excitation rates are zero until the next streamer comes to the surface during a positive cycle of the alternative applied voltage. The electron temperature was chosen as a function of time as follows. Simulation begins with the maximal electron temperature (5 eV) which remains constant for the next 4 µs. Then in 0.1 µs electron temperature exponentially decreases to the gas temperature (300 K) and remains constant for 76 µs until the next positive cycle. After that electron temperature exponentially increases to the maximum value of 5 eV in 0.1 µs.
Due to presence of the sheath near the surface the ion fluxes were added in the model. Ions’ velocity is considered to be equal to Bohm velocity, electrons’ flux equals to the sum of ion’s fluxes to satisfy the quasi-neutrality condition. Also it is considered that all ions recombine after their contact with surface and return in the form of corresponding neutrals. The evolution of concentration of species is described with the following equations

$$\frac{\partial n_\alpha}{\partial t} = \sum_{\lambda=1}^{R_\alpha} \left( v_\lambda^R - v_\lambda^L \right) k_\lambda \prod_{i=1}^{M} n_i^L = \Gamma_\alpha$$

where $n_\alpha$ is number density of $\alpha$th component, $v_\lambda^R$ and $v_\lambda^L$ are right- and left-handed stochastic coefficients of $\lambda$th reaction, $N_\alpha$ is total number of reactions where $\alpha$th component was participated, $k_\lambda$ is rate coefficient of $\lambda$th reaction, $\Gamma_\alpha$ is the flux of $\alpha$th component (positive for ions and electrons, negative for neutral components obtained after ions’ recombination, zero for others), $d$ is estimated thickness of boundary layer (~0.1 mm).

The initial values of $n_{He}, n_{O_2}, n_{N_2}$ and $n_{H_2O}$ were taken from the solution of Navier-Stokes equations for the point corresponding to the point of streamer contact with surface. In our case they equal to $He/N_2/O_2/H_2O = 0.9767/0.0176/0.0050/0.0007$.

Figure 7. Evolution of some radicals after 20 pulses of electron temperature.

Figure 7 shows the evolution of several species for long-time scale (about 16 minutes) after 20 pulses. Concentrations of $H_2O_2$, $NO$, $HNO_x$, $NO_2$, $O_2(a)$, $O_3$ remain high. Concentrations of $OH$ sharply decreases at first ~0.01 s as $OH$ is converting into $H_2O_2$. $O_3$, $N_2O_5$ and $HNO_4$ decays for 1-10 s. $HNO_2$, $H_2O_2$, $NO$ and some other radicals stay constant at this time scale.

5. Conclusion

In this work streamer’s interaction with conductive and dielectric surfaces and effect of external electrode were studied. The rate of ionization and electric field distribution near the surface were calculated using 2D modelling. Two main stages the ionization rate’s evolution were distinguished – the stage of fast intensive ionization (~100 ns) and the stage of slow equilibrium ionization (~10 μs). Obtained results are in the agreement with the experimental observations. It was found that in case with negatively biased external electrode the rate of ionization is higher by factor of 2 and electric field is larger by 30% than that in case without external electrode. Also electric field was much higher over the dielectric surface compared to the conductive one due to presence of surface charge. 0D plasma chemistry calculation was also conducted where the temperature of electrons was taken as a
periodical function of the time. The evolution of concentrations of RONS were calculated in the framework of 0D chemical model.

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